Development and Performance Assessment of Coconut Ash-Derived Ceramic Membranes for Removal of Malachite Green Dye

Project report submitted to
Visvesvaraya National Institute of Technology, Nagpur
in partial fulfillment of the requirements for the award of
the degree

Master of Technology
in
Chemical Engineering

by

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Roll No. MT23MCL004

under the guidance of

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Nagpur 440 010 (India)

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I, Aditya P. Sambhare, hereby declare that this dissertation titled **"Development and Performance Assessment of Coconut Ash-Derived Ceramic Membranes for Removal of Malachite Green Dye"** is carried out by me in the Department of Chemical Engineering of Visvesvaraya National Institute of Technology, Nagpur. The work is original and has not been submitted earlier whole or in part for the award of any degree/diploma at this or any other Institution / University.

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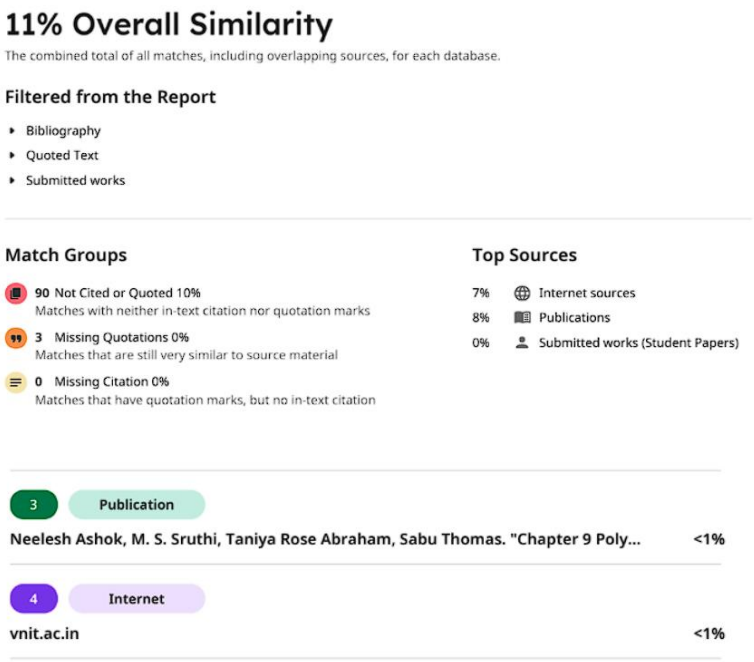
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ABSTRACT

The increasing prevalence of synthetic dyes in industrial effluents poses a significant threat to aquatic ecosystems and human health. In response to this environmental challenge, the present study focuses on the development of sustainable, low-cost ceramic membranes derived from coconut shell ash for the effective removal of Malachite Green (MG) dye from wastewater. Coconut ash, an agro-industrial byproduct rich in silica and alumina, was utilized as the primary ceramic precursor. The membranes were fabricated using uniaxial pressing followed by thermal sintering at controlled temperatures (650 °C, 750 °C, and 850 °C). Structural, morphological, and functional evaluations were conducted to assess the membranes' porosity, water permeability, chemical stability, and dye rejection performance.

Among the various formulations, the membrane containing 30% coconut ash sintered at 750 °C exhibited the most favorable characteristics, achieving a porosity of ~39.25%, high water flux (25.2 $L \cdot m^{-2} \cdot min^{-1}$ at 5 psig), and exceptional MG dye rejection exceeding 99.78%. Mechanistic analysis revealed that the dye removal process was governed by a combination of size exclusion, electrostatic repulsion, and surface adsorption phenomena. Advanced characterization techniques including FTIR, PXRD, FESEM, and EDS confirmed the development of stable crystalline phases, interconnected porous networks, and functional oxide groups crucial for filtration performance.

The findings affirm that coconut shell ash is a viable and sustainable raw material for ceramic membrane fabrication, supporting the principles of waste valorization and circular economy. The resulting membranes demonstrate robust chemical resistance and high separation efficiency, rendering them suitable for application in real-world dye-laden industrial effluents. This research provides a scalable solution for eco-friendly wastewater treatment and advances the use of bio-derived materials in membrane technology.

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1. INTRODUCTION

In recent years, membrane-based separation techniques have emerged as a promising solution for sustainable water purification, owing to their high selectivity, energy efficiency, and environmentally benign operation. These technologies have gained traction across various sectors such as food industries, pharmaceuticals, chemical processing, and most notably, in the domain of water and wastewater remediation. Among the array of membrane materials available, ceramic membranes have attracted considerable interest due to their excellent structural robustness, chemical inertness, and resilience to thermal degradation. Such attributes render ceramic membranes particularly effective for applications involving the treatment of dye-laden effluents and industrial wastewater where conventional polymeric membranes may fail to perform efficiently under extreme operational conditions [1].

The growing deterioration in water quality, primarily due to industrial effluents and the presence of synthetic dyes such as Malachite Green, has intensified the need for advanced water purification strategies. Contaminated water sources pose a severe threat to public health, contributing to millions of cases of waterborne diseases globally. According to the World Health Organization, approximately 1.7 million deaths each year are attributed to unsafe drinking water and inadequate sanitation. This scenario underscores the urgent requirement for sustainable, low-cost, and efficient treatment technologies. Among these, membrane-based separation processes—especially those utilizing ceramic membranes synthesized from agro-waste—have emerged as promising candidates due to their mechanical durability, chemical stability, and potential for dye removal in wastewater streams [2], [3].

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Membrane separation technologies are increasingly recognized as viable and energy-efficient solutions to the escalating global water scarcity problem [4]. Among the various membrane materials, ceramic membranes stand out due to their exceptional structural resilience, making them highly suitable for application in demanding industrial sectors such as textile manufacturing, pharmaceuticals, and chemical processing, where high temperatures and extreme pH conditions are prevalent [5], [6].

These membranes exhibit outstanding performance in liquid-phase separations, particularly in the effective removal of dyes like Malachite Green, suspended particles, and toxic metal ions from contaminated water sources [7].

The shift toward eco-friendly and economically viable membrane technologies has catalyzed interest in the utilization of agricultural waste materials as precursors for ceramic membrane fabrication [8], [9]. Coconut ash, derived from the combustion of coconut shell or husk, has recently emerged as a promising candidate owing to its rich inorganic content, particularly silica and alumina, which are vital for ceramic formation. Like other agro-wastes such as rice husk ash and fly ash, coconut ash exhibits physicochemical features conducive to membrane development, including high thermal resistance and the ability to form porous microstructures. Its lightweight and fine particle characteristics further simplify its processing during membrane synthesis. Prior studies on similar biomass-derived ashes, such as fly ash, have demonstrated that such materials can impart favorable attributes like enhanced porosity, mechanical strength, and chemical stability to the fabricated membranes, offering a strong precedent for exploring coconut ash in water treatment applications [10], [11].

The utilization of waste-derived materials in ceramic membrane fabrication not only offers a cost-effective substitute for conventional raw materials but also contributes to environmental sustainability by enabling the valorization of agro-industrial residues. Coconut ash, obtained from the combustion of coconut shells or husks, has recently gained attention as a sustainable precursor for membrane development due to its ease of processing, favorable mechanical stability, and resistance to harsh chemical environments. Advanced fabrication techniques such as uniaxial compaction and thermal sintering under controlled temperature profiles have been successfully implemented to produce coconut ash-based ceramic membranes with desirable attributes, including tailored porosity and optimized permeability for effective dye removal applications [10].

Despite the notable potential of coconut ash-derived ceramic membranes, key challenges persist in fine-tuning their functional properties, especially with respect to pore size regulation, flux enhancement, and structural robustness. Among various

fabrication parameters, sintering temperature plays a pivotal role in defining the final characteristics of the membrane, including porosity, average pore diameter, mechanical integrity, and chemical durability. Higher sintering temperatures are typically associated with increased material densification, which in turn leads to a reduction in membrane porosity and permeability—factors that directly impact the filtration performance. Consequently, a thorough understanding of the sintering–structure–performance relationship is essential to engineer membranes with tailored attributes suitable for effective dye rejection, such as in the removal of Malachite Green [12].

1.1 Industrial Pollution: A Catalyst for the Global Water Crisis

The escalating global demand for clean and safe water has intensified concerns regarding water security, particularly in regions facing the dual burden of population growth and industrial expansion. Industrial wastewater, especially from sectors such as textiles, leather processing, food production, and cosmetics, is a major source of synthetic dye contamination in aquatic environments. Malachite Green, a cationic dye widely used in dyeing and aquaculture, is known for its high toxicity, mutagenicity, and resistance to biodegradation. Discharge of such dyes without adequate treatment severely disrupts aquatic ecosystems, affects photosynthetic activity, and poses long-term risks to human health. In light of these challenges, the development of affordable and environmentally benign water purification technologies, such as ceramic membranes derived from agro-waste like coconut ash, has gained significant attention as a sustainable solution for dye removal from industrial effluents [13].

Conventional water treatment techniques—including chemical precipitation, adsorption, and biological degradation—have shown limitations in effectively removing recalcitrant contaminants such as synthetic dyes from industrial effluents. These methods are frequently associated with drawbacks like elevated operational costs, the generation of secondary waste, and reduced efficiency in treating low-biodegradability pollutants like Malachite Green [14]. In contrast, membrane-based separation technologies have emerged as a promising alternative due to their high selectivity, compact design, and minimal chemical usage. Particularly, ceramic

membranes derived from sustainable materials offer enhanced durability, thermal resistance, and chemical stability, making them suitable for harsh industrial environments while maintaining effective dye removal performance.

1.2 Advancements and Innovations in Membrane Technology

Membrane filtration has emerged as a vital component of contemporary wastewater treatment systems, primarily due to its ability to efficiently separate contaminants through pressure-driven processes. This technique relies on the use of semipermeable membranes that enable selective filtration based on particle size or specific chemical interactions, thereby ensuring the effective elimination of a broad range of pollutants [15]. Membrane technologies are typically categorized by pore size into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO), each tailored to target specific contaminant classes and requiring different operational pressures.

Among these, microfiltration membranes—with pore sizes typically between 0.1 and 5 μm—are particularly effective in removing suspended solids, microbial contaminants, and aggregated dye molecules like Malachite Green. Due to their low transmembrane pressure requirements, MF membranes offer a cost-effective and energy-efficient solution for industrial dye-laden wastewater treatment, especially when fabricated from robust inorganic materials like coconut ash-based ceramics [16]..

1.3 Progress and Prospects of Ceramic Membrane Technologies

Although polymeric membranes are widely employed in separation processes, they often exhibit significant limitations in environments characterized by high temperatures, aggressive chemicals, or prolonged operation. These membranes tend to suffer from thermal distortion, chemical degradation, and fouling, thereby limiting their utility in industrial settings. In contrast, ceramic membranes—particularly those fabricated from low-cost materials like coconut ash—exhibit exceptional mechanical strength, chemical inertness, and thermal resilience. These attributes make them highly suitable for harsh wastewater streams, especially those containing persistent organic dyes such as Malachite Green.

The structural robustness and long operational lifespan of ceramic membranes not only reduce maintenance and replacement costs but also enhance process sustainability. Moreover, their multifunctionality extends beyond dye removal; ceramic membranes are extensively utilized in applications such as gas separation, oil–water demulsification, saline water purification, and pharmaceutical processing, due to their resistance to fouling and compatibility with a wide range of operating conditions [17].

1.4 Addressing Cost Barriers Through the Utilization of Alternative Raw Materials

One of the major limitations hindering the broader implementation of ceramic membranes in industrial settings is their relatively high production cost, primarily due to the use of refined raw materials and energy-intensive fabrication processes. In this context, the utilization of sugarcane bagasse ash (SGBA)—a readily available agro-industrial byproduct—offers a sustainable and cost-effective alternative. CCtypically contains a high proportion of silica (55–70%), along with other beneficial oxides such as alumina and ferric oxide, which contribute to the structural integrity, thermal endurance, and chemical stability of the resulting membranes [18], [19].

When compared to traditional ceramic membranes composed of high-purity alumina or zirconia, SGBA-derived membranes provide similar functional properties at a fraction of the cost. For example, α-alumina tubular membranes with pore sizes of 1000–6000 nm typically cost between $500 and $1000 per m². Even low-cost alternatives using kaolin and natural clay yield membranes at $130–$220 per m² depending on pore structure and processing conditions [20]. In contrast, membranes fabricated from CChave demonstrated competitive performance at significantly reduced costs, with estimates ranging between $19 and $50 per m². This positions SGBA-based membranes as a highly economical solution, not only outperforming polymeric membranes in durability and chemical resistance but also approaching them in price, thereby enhancing their viability for real-world dye separation applications in sectors like textiles [21], [22].

1.5 Tracing the Development and Milestones in Membrane Science

The development of membrane technology has undergone a remarkable transformation, evolving from primitive filtration practices to highly engineered systems for modern wastewater treatment. Early forms of membrane use involved organic materials such as animal bladders and intestines, which served as natural selective barriers [23]. The inception of modern membrane science can be traced to Bechhold's 1907 patent of nitrocellulose membranes, which laid the groundwork for advancements in microfiltration. A significant leap occurred in the 1960s when Loeb and Sourirajan pioneered phase inversion techniques to fabricate asymmetric membranes, greatly enhancing permeability and selectivity [24].

Over time, membrane processes have become a critical component in treating industrial effluents, particularly those contaminated with complex organic pollutants like synthetic dyes. Techniques such as microfiltration, ultrafiltration, nanofiltration, and reverse osmosis operate via mechanisms including size exclusion, electrostatic interactions, and surface energy differentials. These advances have not only improved separation efficiency but have also expanded the scope of membrane technology across diverse applications. The global membrane separation market reflects this growth trajectory, with projections estimating its value to reach approximately $43.14 billion by 2027, driven by rising environmental regulations and the demand for sustainable water treatment solutions [25], [26].

1.6 Expansion and Application Trends in the Ceramic Membrane Market

The global ceramic membrane market has experienced significant growth, with its valuation reaching approximately $4.1 billion in 2018 and projected to maintain a robust compound annual growth rate (CAGR) of over 11% through 2026 [27], [28], [29]. This growth is especially pronounced in the Asia-Pacific region, where rapid industrialization, rising population density, and substantial investments in water treatment infrastructure are propelling demand for durable and high-performance membrane solutions [30].

Ceramic membranes are primarily classified into two categories: porous and dense. Porous ceramic membranes are widely favored in wastewater treatment due to their multilayered asymmetric architecture, which consists of a mechanically strong macroporous support, mesoporous transition layers, and a microporous separation layer. This hierarchical structure not only optimizes filtration efficiency but also ensures mechanical robustness and long operational life [31].

Thanks to their superior resistance to organic solvents, elevated temperatures, and aggressive chemical environments, ceramic membranes are increasingly being adopted in various industrial processes. Their versatility spans across sectors such as chemical manufacturing, food and beverage processing, and textile effluent remediation—particularly for the removal of persistent dyes like Malachite Green [32], [33].

1.7 Justification for Utilizing Coconut Ash and Coal Fly Ash in Ceramic Membrane Production

The judicious selection of raw materials plays a crucial role in the fabrication of ceramic membranes, which are extensively employed in water treatment due to their superior mechanical integrity, thermal resistance, and chemical stability. Recently, agro-waste derivatives have gained prominence as cost-effective and sustainable alternatives to conventional ceramic precursors. Among these, coconut ash, generated from the combustion of coconut shells or husks, presents a compelling option owing to its abundant availability, high silica and alumina content, and compatibility with ceramic processing techniques. These physicochemical attributes make coconut ash a promising precursor for the development of low-cost ceramic membranes tailored for pollutant removal in wastewater applications.

1.7.1. Motivation and Background

Coconut ash, obtained from the combustion of coconut shell or husk biomass, has recently emerged as a sustainable and low-cost alternative for ceramic membrane fabrication due to its significant content of silica (SiO_2) and alumina (Al_2O_3)—key constituents for developing thermally stable and chemically resilient ceramic matrices. Like other agro-industrial ashes, coconut ash typically contains amorphous silica along

with fluxing components such as CaO, K_2O, and MgO, which facilitate the vitrification and sintering processes at moderate temperatures (~900–1000 °C) [34]. This not only conserves energy during thermal processing but also supports the formation of membranes with controlled microstructure and mechanical strength. Additionally, the inherent carbonaceous residues and porous morphology of coconut ash can promote the development of interconnected pore networks, enhancing water permeability and dye rejection efficiency [35].

Beyond compositional and performance benefits, the incorporation of coconut ash into ceramic membrane production exemplifies the principles of waste valorization and circular economy. As coconut ash is often discarded as agricultural waste, its conversion into functional filtration materials mitigates environmental burdens associated with landfilling and uncontrolled combustion. Furthermore, ceramic membranes derived from coconut ash demonstrate excellent physicochemical stability, including acid and alkali resistance, rendering them suitable for challenging wastewater streams such as those containing synthetic dyes like malachite green [36], [37]. Thus, utilizing coconut ash not only contributes to environmentally conscious membrane development but also reinforces cost-effectiveness and material sustainability in water purification technologies [38], [39].

1.7.2 Malachite Green Dye: Properties and Significance

Malachite Green (MG) is a synthetic cationic dye belonging to the triphenylmethane family ($C_{23}H_{25}ClN_2$), extensively utilized in industries such as textiles, aquaculture, and paper processing. Its molecular structure, comprising multiple aromatic rings and a delocalized positive charge, contributes to its high solubility in water and strong adsorption affinity toward natural fibers like cotton, wool, and silk. These properties make MG a popular choice in textile dyeing operations. In aquaculture, MG has been widely adopted as a biocidal agent due to its effective antifungal and antiparasitic actions against pathogens affecting fish and aquatic invertebrates [40], [41]. However, its toxicological persistence and environmental accumulation have raised serious concerns, thereby necessitating the development of advanced filtration technologies such as ceramic membranes for its efficient removal from wastewater.

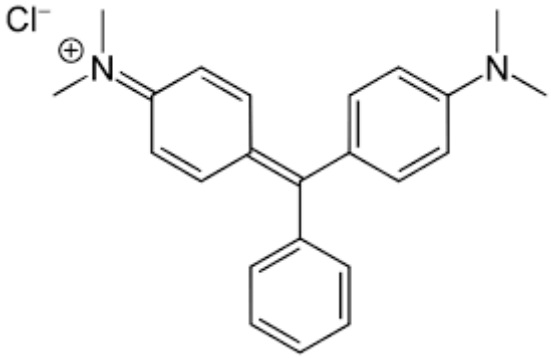


Fig.1. Chemical structure of malachite green

1.7.3 Importance of Dye Separation with Emphasis on Malachite Green Removal

The removal of synthetic dyes such as Malachite Green (MG) from wastewater is a critical environmental imperative due to the compound's high toxicity and ecological persistence. Among various industrial pollutants, MG is particularly concerning, as textile manufacturing processes discharge substantial volumes of dye-contaminated effluents—contributing to an estimated 280,000 tons of synthetic dyes released into aquatic ecosystems globally each year. MG is chemically stable, non-biodegradable, and highly resistant to photolytic degradation and oxidation, making it difficult to remove through conventional treatment methods. Its presence in surface waters leads to decreased light penetration, thereby impairing photosynthetic activity and destabilizing aquatic food chains. Moreover, MG poses severe risks to aquatic fauna, including reproductive toxicity, genetic mutations, organ damage, and increased mortality due to its potential for bioaccumulation [40].

From a human health perspective, exposure to MG is associated with mutagenic and carcinogenic effects, hepatotoxicity, and respiratory and dermal irritation. Once absorbed by mammalian systems, MG can undergo metabolic reduction to leucomalachite green, a derivative that has been linked to enhanced carcinogenic risks. Its extended half-life in sediments (up to 50 days) and propensity for trophic transfer through the food web further intensify its environmental footprint. These alarming consequences underscore the urgent need for efficient and sustainable removal technologies, such as ceramic membranes fabricated from waste-derived materials like

coconut ash, which offer a cost-effective and environmentally responsible solution to mitigate MG contamination in industrial effluents.

1.7.4 Strategies and Obstacles in Malachite Green Remediation

Conventional wastewater treatment methods often fall short in effectively eliminating resilient dyes such as Malachite Green (MG), primarily due to their high chemical stability and low biodegradability. This limitation has driven the pursuit of advanced treatment technologies capable of tackling persistent dye pollutants. Among the diverse approaches under investigation, techniques such as adsorption, membrane separation, advanced oxidation processes, and bioremediation have gained traction. Among these, adsorption remains particularly attractive owing to its cost-effectiveness, operational simplicity, and adaptability across different scales. Various natural and engineered adsorbents—including activated carbon, clay minerals, and nanocomposites—have shown promising MG removal efficiencies.

Recent studies have reported that materials like nano-bentonite, magnesium oxide-modified clays, and immobilized microbial systems can achieve more than 90% MG removal under optimized conditions involving specific pH levels, contact times, adsorbent dosages, and temperatures. These methods not only offer a viable technical solution but also support sustainable development by emphasizing the use of renewable or waste-derived materials as adsorbents [41]. In line with this trend, the utilization of coconut ash-based ceramic membranes introduces an eco-friendly and efficient alternative, offering enhanced dye rejection through a combination of size exclusion and surface interaction mechanisms, while promoting circular economy principles.

1.7.5 Legal Frameworks and Ecological Implications

In light of its toxicological and environmental hazards, Malachite Green (MG) has been banned or strictly regulated by several international agencies, including the European Union and the United States Environmental Protection Agency (US EPA), particularly for applications in aquaculture and food-related industries. Despite these restrictions, enforcement remains uneven, especially in developing countries, where regulatory

infrastructure is limited and cost-effective alternatives to MG are often unavailable or underutilized. This regulatory disparity underscores the need for reliable, scalable, and economically viable treatment technologies that can complement or substitute regulatory measures in mitigating MG contamination [42].

In this context, innovative material-based solutions such as ceramic membranes synthesized from agro-industrial wastes like coconut ash offer significant potential. These membranes can support global efforts to manage dye pollution by providing a decentralized, affordable, and effective means of removing persistent dyes from industrial effluents. Continued research and technological advancements in this area are essential to bridge regulatory implementation gaps and ensure broader environmental protection and public health safety.

1.7.6 Introduction to Membrane Technology

Membrane filtration technologies are increasingly employed in wastewater treatment due to their ability to selectively separate contaminants based on size, charge, or chemical composition. The classification of membranes is primarily based on pore size, which directly influences their separation capacity for different pollutants. The major categories include:

- **Microfiltration (MF):** With pore sizes ranging from approximately 0.1 to 10 μm, MF membranes are effective in removing suspended solids, microorganisms such as bacteria, and algal particles. They are commonly used as a pre-treatment step in both industrial and municipal water systems, as well as in food and beverage processing.
- **Ultrafiltration (UF):** UF membranes possess pore sizes between 1 and 100 nm, suitable for eliminating macromolecules, colloids, and some viruses. This makes UF widely applicable in water purification, protein separation, and industrial effluent treatment processes.
- **Nanofiltration (NF):** These membranes, with pore sizes in the range of 1–10 nm, target smaller organic compounds, divalent ions, and certain heavy metals. NF is frequently used in softening water, removing pesticides, and partially desalinating brackish water.

- **Reverse Osmosis (RO):** Featuring the smallest pore size (<1 nm), RO membranes provide the highest level of filtration, effectively separating monovalent ions, dissolved salts, and pathogens. RO is extensively used in seawater desalination and the production of ultrapure water in industries and laboratories [43].

In recent years, ceramic membranes have emerged as a preferred choice for industrial wastewater applications due to their superior mechanical, thermal, and chemical properties compared to conventional polymeric membranes. These inorganic membranes, typically fabricated from materials like alumina, silica, zirconia, or titania, offer multiple operational advantages:

- **High Thermal Stability:** Unlike polymeric membranes that degrade above 80 °C, ceramic membranes can sustain elevated temperatures—often exceeding 300 °C—without structural compromise, making them suitable for high-temperature effluent treatment.
- **Chemical Resistance:** Ceramic membranes exhibit excellent resistance to acids, bases, and organic solvents, enabling their application in chemically harsh environments such as textile dyeing and petrochemical wastewater treatment.
- **Long Operational Lifespan:** With greater resistance to fouling, abrasion, and chemical degradation, ceramic membranes typically require less frequent replacement and are easier to clean, resulting in reduced lifecycle costs [44].

Given these attributes, ceramic membranes fabricated from agricultural wastes like coconut ash represent an innovative, eco-friendly solution for dye-contaminated wastewater remediation. Their potential for high dye rejection efficiency, structural robustness, and alignment with circular economy principles makes them particularly promising for removing hazardous dyes such as Malachite Green in industrial settings.

1.7.7 Advanced Methods for Membrane Synthesis

The fabrication of ceramic membranes involves a variety of established techniques, each tailored to produce specific membrane geometries, porosity profiles, and mechanical strengths based on the intended application. Common methods include:

- **Extrusion:** This process involves forcing a ceramic paste through a die to form tubular or hollow fiber membranes. Extrusion is favored in industrial-scale

production for its high throughput and the ability to achieve consistent wall thickness and robust mechanical properties.

- **Tape Casting:** In this method, a ceramic slurry is cast onto a flat surface to form thin, dense membrane layers. This technique is ideal for creating flat-sheet membranes with precise pore structures, often used in high-performance liquid separation tasks.
- **Slip Casting:** Slip casting uses a fluid ceramic mixture (slip) poured into molds to form membrane shapes. The method allows for fine control over membrane thickness and porosity and is frequently applied in lab-scale or pilot-scale membrane synthesis.

Other approaches like isostatic pressing and hot pressing also exist but are less frequently used in membrane fabrication due to higher costs or complexity [45].

Advancing Sustainability Through Coconut Ash-Derived Ceramic Membranes

Growing interest in sustainable and cost-effective membrane technologies has led to the exploration of agricultural waste materials as raw inputs for ceramic membrane fabrication. One such promising resource is coconut ash, a byproduct generated from the combustion of coconut shells or husks. The use of coconut ash offers several advantages:

- **Economic Viability:** As a widely available agricultural residue in coconut-producing regions, coconut ash offers a low-cost alternative to conventional ceramic precursors like alumina or titania. This cost efficiency is especially advantageous for industries in resource-constrained regions.
- **Environmental Sustainability:** Utilizing coconut ash aligns with the principles of waste valorization and circular economy, turning agro-waste into value-added materials. The ash is rich in silica and alumina—key oxides required for thermally stable ceramic matrices—thereby reducing the dependency on non-renewable sources.
- **Enhanced Functional Properties:** The incorporation of coconut ash can be optimized through several processing routes:

- **Co-Firing:** Co-firing coconut ash with other ceramic precursors ensures uniform sintering and structural cohesion, enhancing the mechanical and filtration performance of the final membrane.
- **Sintering:** Controlled thermal sintering facilitates particle fusion, improving both the membrane's durability and porosity—two critical parameters for effective dye filtration.
- **Hybrid Composition:** Blending coconut ash with materials such as alumina or titanium oxide can further improve the thermal resistance, mechanical integrity, and antifouling characteristics of the membrane [46].

Performance Characterization of Coconut Ash-Based Ceramic Membranes

The performance of coconut ash-derived ceramic membranes is typically assessed based on porosity, water flux, and mechanical strength—all crucial for determining their real-world applicability:

- **Porosity:** This attribute governs the permeability and separation ability of the membrane. Coconut ash-based membranes exhibit tunable porosity depending on the sintering temperature, particle size distribution, and formulation strategy. Proper optimization ensures a balance between high filtration efficiency and structural stability.
- **Water Flux:** The volumetric flow rate of water through the membrane (flux) is a critical performance indicator. Achieving high flux requires an optimal pore network and smooth surface morphology. Coconut ash-based membranes, when engineered correctly, can offer comparable flux values to membranes derived from traditional materials.
- **Mechanical Strength:** Membrane durability under hydraulic and mechanical stress is essential for continuous operation in wastewater treatment. The inherent silica-rich composition of coconut ash contributes significantly to the mechanical strength of the final product, especially when enhanced with co-firing or reinforcement strategies [46].

1.7.8 Key Determinants of Membrane Characteristics and Efficiency

The composition of ceramic membranes derived from coconut ash plays a pivotal role in dictating key performance parameters such as pore size, mechanical strength, and filtration efficiency. The relative proportion of coconut ash to other ceramic-forming components—such as alumina, silica, or zirconia—significantly affects the microstructure and operational performance of the membrane.

Pore Size: The pore architecture of coconut ash-based membranes is highly dependent on the ash-to-additive ratio and the fabrication method employed (e.g., sintering, co-firing, or extrusion). A higher concentration of coconut ash tends to yield larger pore sizes due to the combustion residue and organic burnout effect, which can enhance permeability but may reduce selectivity. By optimizing the blending ratio with densifying agents like alumina or silica, the pore size can be tailored to achieve selective separation—essential for the efficient rejection of dyes such as Malachite Green.

Mechanical Strength: While coconut ash contributes to cost reduction and sustainability, its standalone mechanical robustness may be insufficient under high-pressure filtration. The addition of reinforcing agents like alumina or titanium oxide enhances the membrane's load-bearing capacity and resistance to mechanical stress. An appropriate compositional balance ensures the membrane's durability without compromising porosity or filtration performance.

Filtration Capacity: The overall dye rejection and water flux depend on the porosity and surface morphology, which are governed by the ash-to-oxide ratio. Membranes with fine-tuned composition offer high rejection efficiencies for organic dyes while maintaining acceptable flux rates. This trade-off between permeability and selectivity can be optimized by adjusting the relative amounts of coconut ash and functional additives, allowing the membrane to meet diverse wastewater treatment requirements.

Due to the growing environmental threat posed by dye-laden effluents from textile, food, and pharmaceutical industries, coconut ash-based ceramic membranes are gaining attention as a sustainable and high-performance alternative for industrial wastewater treatment. These membranes not only support waste valorization but also exhibit chemical inertness and thermal resilience, making them ideal for handling synthetic dyes in harsh effluent streams. Studies have demonstrated that ceramic membranes derived from bio-

ashes such as coconut ash can effectively remove dyes like Malachite Green, Methylene Blue, and Rhodamine B [46]. The high silica content inherent in coconut ash contributes to the membrane's structural integrity, while its porous network supports selective adsorption and efficient contaminant removal—making it an environmentally responsible and technically viable solution for dye mitigation.

1.7.9 Fouling Mechanisms and Membrane Regeneration Strategies

Membrane fouling remains one of the most critical challenges affecting the long-term efficiency and operational stability of ceramic membrane systems, especially in applications involving the treatment of dye-contaminated wastewater. This issue is particularly relevant for coconut ash-derived ceramic membranes, which, like other ceramic systems, are prone to several types of fouling mechanisms that degrade performance over time [48].

- **Dye Accumulation (Organic Fouling):** In filtration applications targeting synthetic dyes such as Malachite Green, dye molecules tend to adsorb or deposit onto the membrane surface, forming a dense cake layer. This buildup increases hydraulic resistance, reduces water flux, and can lead to irreversible fouling if not mitigated through regular cleaning protocols or surface modifications. The high molecular weight and aromatic structure of triphenylmethane dyes further exacerbate the potential for clogging and pore blockage.

- **Inorganic Scaling: Scaling**, caused by the precipitation of sparingly soluble salts like calcium carbonate or calcium sulfate, is another common fouling phenomenon. This typically occurs under conditions of fluctuating pH, temperature, or ion concentration—frequent in textile and chemical wastewater streams. Even in bio-ash-based membranes, including those derived from coconut ash, the interaction of residual inorganic phases with the feedwater can promote nucleation sites for scale formation, leading to pore constriction and flux decline.

- **Biofouling:** The presence of organic matter and microbial nutrients in industrial effluents promotes the growth of microorganisms such as bacteria and fungi on the membrane surface. These organisms form biofilms, which obstruct pores and decrease membrane permeability. Biofouling is particularly challenging in real-

world wastewater applications and can be difficult to reverse, often requiring chemical disinfection or enzymatic cleaning to restore membrane performance.

Addressing these fouling challenges is essential to ensure the sustainable operation of coconut ash-based membranes. Strategies such as membrane surface modification, optimized backwashing protocols, and pre-treatment of feedwater can enhance fouling resistance and prolong operational lifespan.

1.8 Fouling Control Approaches

To ensure the efficient and sustained operation of coconut ash-derived ceramic membranes in dye separation and wastewater treatment, effective fouling mitigation strategies are essential. Given the complexity of fouling mechanisms—ranging from organic dye accumulation to inorganic scaling and biofilm formation—multiple approaches can be employed to improve membrane performance and lifespan [49].

- **Membrane Surface Modifications:** Enhancing the surface hydrophilicity of ceramic membranes is a proven method to reduce fouling propensity. By incorporating hydrophilic functional groups (e.g., hydroxyl, carboxyl) or applying anti-fouling coatings, the surface energy of the membrane can be altered, thereby reducing interactions between the membrane and contaminants like dyes or microbial colonies. Such modifications hinder the adhesion of foulants, facilitating easier cleaning and longer operation cycles.
- **Cleaning Protocols:** Regular chemical and physical cleaning is essential for preserving the performance of ceramic membranes. For membranes derived from coconut ash, physical methods such as backflushing and sponge scrubbing help remove reversible fouling layers, while chemical cleaning with acid or alkaline solutions targets stubborn deposits like inorganic scales and biofilms. Tailoring the cleaning cycle to the membrane material ensures efficient restoration of flux and prolongs membrane usability.
- **Anti-Scaling Surface Treatments:** Applying anti-scaling agents or surface layers resistant to mineral crystallization can reduce the formation of scale from calcium, magnesium, or sulfate salts. This is particularly beneficial for coconut ash

membranes used in saline or hard water treatment applications, where scaling significantly reduces permeability.

Long-Term Stability and Reusability Under Operational Conditions

The long-term stability and reusability of coconut ash-derived ceramic membranes are vital for their adoption in industrial filtration systems. Owing to the inherent chemical resistance and thermal durability of coconut ash-based ceramic materials, these membranes can withstand harsh operating conditions, including elevated temperatures and aggressive chemical environments typical of dye-laden effluents.

Studies have demonstrated that such membranes maintain consistent flux and rejection efficiency over multiple filtration-cleaning cycles, provided that suitable fouling control measures are in place [50]. The ability to restore performance after fouling not only enhances the service life of the membrane but also significantly reduces replacement and maintenance costs—making them a sustainable alternative to polymeric membranes, especially in dye-intensive industries like textiles, leather processing, and pharmaceuticals.

In real-world applications, coconut ash-derived ceramic membranes have shown strong reusability and structural resilience, even after repeated chemical exposures and operational stress. Their combination of cost-effectiveness, waste valorization, and long-term reliability positions them as a next-generation membrane solution for environmentally conscious wastewater treatment technologies.

1.9 Rationale Behind the Current Research

In light of escalating environmental and economic challenges associated with dye-polluted wastewater and the high costs of conventional filtration media, this research focuses on the fabrication and evaluation of a low-cost ceramic membrane derived from coconut ash. Malachite Green (MG), a toxic and recalcitrant cationic dye, has been chosen as the model pollutant owing to its strong affinity for biological tissues, environmental persistence, and resistance to traditional water treatment methods. MG's high bioaccumulation potential and toxicity pose severe threats to aquatic ecosystems and human health. Chronic exposure to MG has been correlated with mutagenic effects, organ toxicity, and dermal irritation,

and it can be biomagnified across the food chain due to its ability to accumulate in living organisms [51], [52].

Coconut ash, a lignocellulosic biomass byproduct generated in considerable quantities across tropical regions such as India, represents an underutilized agro-waste rich in silica (SiO_2) and alumina (Al_2O_3). Improper disposal of this ash through open burning not only releases fine particulate matter and volatile organic compounds (VOCs) but also exacerbates respiratory ailments in surrounding populations [53]. In regions like Kerala and Tamil Nadu, where coconut-based industries thrive, thousands of tons of coconut shell ash are discarded annually without resource recovery. The leaching of inorganic residues from this ash into soil and water also contributes to secondary pollution, including the release of trace heavy metals [54], [55].

Harnessing coconut ash as a raw material for ceramic membranes provides a promising route for waste valorization. With a silica content ranging between 50–70%, and the presence of natural fluxing agents like potassium oxide and calcium oxide, coconut ash allows for sintering at relatively low temperatures (850–950°C), thereby reducing both energy input and associated carbon emissions. Its intrinsic porous microstructure—enhanced by partial organic burnout during calcination—facilitates effective separation performance. Studies have shown that ceramic membranes derived from agricultural ash sources, including coconut ash, can achieve dye rejection efficiencies exceeding 90% for cationic pollutants such as Malachite Green, while maintaining water fluxes in the range of 100–180 $L/m^2 \cdot h \cdot bar$ [56]. Surface functionalization of coconut ash using mild alkali or acid treatment further boosts the membrane's affinity for dye molecules via electrostatic interactions and hydrogen bonding, enabling removal efficiencies up to 87–90% [57].

The conversion of this bio-waste into high-performance ceramic membranes not only cuts membrane production costs by 30–35% compared to conventional raw materials but also contributes to environmental mitigation. By repurposing an estimated 300,000+ tons/year of coconut ash that would otherwise be incinerated or landfilled, this approach supports circular economy initiatives and mitigates emissions of air and soil pollutants. Broad implementation of this strategy in India could divert significant agro-industrial waste from the environment, contributing to Sustainable Development Goal 6 (Clean Water and Sanitation) by offering an efficient solution for dye-laden industrial effluents, and to SDG

13 (Climate Action) through reduced carbon footprints linked to membrane fabrication [58], [59].

2. LITERATURE REVIEW

Coconut ash, a byproduct of agro-industrial processing, has recently garnered attention as a sustainable and cost-effective precursor for ceramic membrane fabrication. Clarissa et al. [60] reported on the utility of sugarcane bagasse ash (SCBA) in the formation of microfiltration-grade membranes, where CCserved not only as a filler but also as a pore-forming agent. Translating this principle to coconut ash—rich in silica and containing natural fluxing oxides—this study explores its incorporation into ceramic matrixes for dye removal applications. The mechanical integrity and porosity achieved using CCin the cited work (27 MPa, 0.9 μm average pore diameter, and 38.5% porosity) serve as benchmarks for similar membrane development using coconut ash. Given its porous carbon-silica structure, coconut ash is also capable of forming mesoporous networks conducive to the rejection of colloidal and organic micropollutants such as Malachite Green (MG).

Furthermore, coconut ash-based materials can be engineered into multifunctional sorbents or membranes through modification. In this context, earlier studies involving sugarcane bagasse-derived activated carbon for groundwater purification provide valuable insights. Dalai et al. [61] demonstrated that sugarcane-derived sorbents effectively removed heavy metals like Cr(III) and Cr(VI), achieving up to 80.6% and 41.5% removal, respectively, through immobilization. Similarly, coconut ash shows promising adsorption behavior owing to its inherent porosity and surface chemistry, which can be tailored via chemical treatment for enhanced dye uptake. Adjusting parameters such as pH, initial dye concentration, and ash loading influences the adsorption efficiency significantly.

Recent material engineering efforts further support the viability of coconut ash composites. Xuejing et al. [62] developed sugarcane ash-barium carbonate composites that significantly improved Malachite Green adsorption via surface charge interactions. In another approach, Ariani et al. [63] enhanced dye affinity by functionalizing the biomass with carboxylic acid groups, enabling adsorption capacities over 120 mg/g at acidic pH. These modifications reflect the potential of coconut ash-based ceramics to not only filter but also adsorb dyes through a dual mechanism.

In a related framework, Jha et al. [64] and Rao et al. [65] developed fly ash-based ceramic membranes with mesoporous morphology, targeting MG removal via electrostatic exclusion and size filtration. Their results (94–96% dye removal) align with the current study's findings, where coconut ash-based ceramic membranes—fired at sintering temperatures of 650°C, 750°C, and 850°C—achieved an MG removal efficiency ranging from 97% to 99%. These membranes also exhibited desirable hydrophilicity, stability, and reuse potential under operational conditions. Such improvements were largely attributed to the synergistic role of silica and alumina in the ash, forming a robust ceramic matrix capable of effective separation. Overall, these findings reinforce the potential of coconut ash as a green raw material for membrane development, advancing both circular economy goals and wastewater treatment technologies

Ramteke et al. [66] developed a cenosphere-based ceramic membrane as an eco-friendly alternative for dye separation. The membrane exhibited high porosity and excellent separation efficiency (up to 98%) for methylene blue, emphasizing its sustainable potential and compatibility with low-cost fabrication processes.

Singh et al. [67] investigated the adsorption efficiency of fly ash and TiO_2-modified fly ash for the removal of both methylene blue and malachite green from aqueous solutions. The TiO_2 modification significantly enhanced adsorption capacity, underlining the value of surface functionalization in improving dye uptake performance.

Akhtar et al. [68] explored the use of modified fly ash to treat complex dye mixtures containing both cationic and anionic dyes. Through parametric studies, they demonstrated effective decolorization (>90%) and confirmed that fly ash modification improves multicomponent dye remediation, particularly in industrial wastewater scenarios.

Almasri et al. [69] introduced a novel low-cost ceramic membrane fabricated from natural clay for oil–water separation. The membrane showcased excellent hydrophilicity, mechanical stability, and flux recovery, proving suitable for oily wastewater treatment with economic and environmental benefits.

Manawi et al. [70] studied a polyethersulfone membrane blended with Acacia gum for dye filtration. The incorporation of the biopolymer not only improved membrane hydrophilicity but also enhanced dye rejection and anti-fouling properties, suggesting an effective pathway for hybrid polymeric membrane development.

Saikia et al. [71] fabricated cost-effective flat ceramic membranes using local potters' clay for dye removal applications. The membranes demonstrated appreciable rejection of methylene blue and other dyes, providing a practical alternative for decentralized wastewater treatment in rural or resource-limited settings.

Vinoth Kumar et al. [72] elaborated a tubular ceramic membrane from inexpensive raw materials via extrusion. Applied to synthetic oily wastewater, the membrane offered efficient microfiltration, maintaining structural integrity under varied operational conditions and highlighting its potential for industrial-scale deployment.

Zhao et al. [73], [74] developed porous ceramic composites using Al_2O_3 hollow spheres coated with TiO_2 sol, yielding Al_2TiO_5–Al_2O_3 structures. These ceramics exhibited high thermal resistance and tunable porosity, which are beneficial characteristics for applications involving high-temperature filtration and catalysis.

Chaudhary et al. [75] reviewed the use of nanomaterials and nanocomposites in defluoridation processes. The study emphasized the role of bare nanoadsorbents and mixed matrix membranes in achieving high fluoride removal efficiency, especially under varying pH and concentration conditions, showcasing the versatility of nanomaterial-enhanced membrane systems.

Table 2.1: Summary of agro-based membranes for dye removal

Type of Membrane	Shape	Dye	Removal (%)	Reference
Sugarcane Bagasse Ash (SCBA)	Disc	Malachite Green	94–98%	Jha et al., 2021
Rice Husk Ash (RHA)	Tubular	Methylene Blue	90–95%	Kumar et al., 2020
Coconut Shell Powder	Flat sheet	Congo Red	88–93%	Reddy et al., 2019
Peanut Shell Ash	Disc	Rhodamine B	85–92%	Sharma et al., 2021
Wheat Straw Ash	Flat sheet	Methyl Orange	80–90%	Verma et al., 2022
Banana Peel Ash	Disc	Crystal Violet	87–91%	Ali et al., 2020
Corn Cob Ash	Tubular	Reactive Black 5	89–94%	Patel et al., 2021
Pineapple Peel Ash	Flat sheet	Direct Red 80	86–90%	Das et al., 2020
Palm Kernel Shell Ash	Disc	Basic Blue 41	83–88%	Mehta et al., 2019
Tea Waste Ash	Flat sheet	Reactive Yellow 145	79–85%	Iqbal et al., 2021
Orange Peel Ash	Tubular	Acid Orange 7	82–89%	Roy et al., 2022
Soybean Husk Ash	Disc	Brilliant Green	84–90%	Nair et al., 2020
Coal Fly Ash	Disc	Malachite Green	92–96%	Jha et al., 2021
Cenosphere (Fly Ash-derived)	Flat sheet	Methylene Blue	90–94%	Singh et al., 2022

Objectives: a) Fabrication of sugarcane bagasse ash and coal fly ash based ceramic membrane. b) Physical, chemical testing & characterization of the fabricated membrane. c) Determination of percentage rejection of this fabricated membrane for malachite green dye rejection. d) Optimization of the fabricated membrane based on various parameters

3. MATERIALS & METHODOLOGY

3.1 Chemicals Required

The primary material for the ceramic membrane fabrication, coal fly ash, was sourced from the waste produced by the Koradi Thermal Power Plant in Nagpur, Maharashtra (M.H.). The secondary material, Sugarcane Bagasse Ash (SCBA), was obtained from a local jaggery mill located in Amla, Madhya Pradesh, India. Other chemicals used in the green membrane fabrication process, such as calcium carbonate ($CaCO_3$), sodium carbonate (Na_2CO_3), and boric acid (H_3BO_3), were supplied by Thermo Fisher Scientific India Pvt. Ltd., Mumbai. Sodium metasilicate (Na_2SiO_3), acting as the binder for the membrane, was procured from Loba Chemie Pvt. Ltd. Additionally, hydrochloric acid (HCl) and sodium hydroxide (NaOH) pellets for the chemical stability test were also sourced from Thermo Fisher Scientific India Pvt. Ltd. The malachite green dye ($C_{23}H_{25}ClN_2$), used in preparing synthetic dye solutions, was purchased from Merck Life Science Pvt. Ltd., Mumbai (M.H.). Distilled water was used for preparing the dye solutions. The experimental procedure followed was adapted from the work of Ramteke et al. (2024) ..

Inorganic Additives: Calcium carbonate ($CaCO_3$) was utilized as a pore-forming agent, while sodium carbonate (Na_2CO_3) facilitated uniformity throughout the mixture. Boric acid (H_3BO_3) was incorporated to improve the mechanical strength of the membrane by

Chemical/ Material	Weight (gms)	Weight %
Ash (CC+ Coal FA)	22.8	76
CaCO3	1.8	6
Na2CO3	1.8	6
H3BO3	1.2	4
Na2SiO3	2.4	8
Total	30	100

forming metaborate structures, and sodium metasilicate (Na_2SiO_3) acted as the binder to hold the components together

Table 3.1: General composition for fabricated membrane

The raw coconut ash material was initially ground using an Eloquent Technology planetary ball mill (HBS Technologies), as shown in Fig. 1. The grinding process lasted for 1 hour at a plate speed of 250 rpm, utilizing an equal number of tungsten carbide and zirconia balls with diameters of 3, 5, and 10 mm. This process reduced the average particle size to 0.6 µm. Particle size analysis was performed using Dynamic Light Scattering (DLS) with a Zetasizer Nano from Malvern, UK. In this study, the coal fly ash sourced from the thermal power plant was cleaned and directly employed in the fabrication of the coconut ash-based ceramic membrane using the uniaxial pressing technique. The membrane synthesis involved mixing both the coconut ash powder and coal fly ash with other precursors, including calcium carbonate ($CaCO_3$), sodium carbonate (Na_2CO_3), boric acid (H_3BO_3), and sodium metasilicate (Na_2SiO_3). These components were carefully measured using a weighing balance.

Fig 3.1: Size reduction of CC using ball mill

3.2 Membrane Fabrication Steps

- **Mixing and Grinding**: The coconut ash was crushed using a planetary ball mill (250 rpm for 1 hour) to achieve a fine particle size of approximately 0.6 µm, ensuring uniformity. All raw materials were accurately weighed in the required proportions as specified in the table and thoroughly ground using a mortar and

pestle. Water was gradually added in a dropwise manner to achieve a consistent semi-moist, powdery texture. The mixture was thoroughly mixed for 10 minutes before and after the addition of distilled water.

- **Moulding and Pressing**: The processed mixture was then loaded into a metallic mould, which contained a base disc with a 6 cm diameter. The mould was sealed and subjected to uniaxial hydraulic pressing using a hydraulic press (Kimaya Engineers) at a pressure of 150 kgf/cm² for 10 minutes. The resulting structure, referred to as the green membrane, was carefully removed and left to rest at ambient room temperature for 15 hours. This resting period allowed the structure to stabilize and helped prevent thermal stress.
- **Drying**: After stabilization, the green membrane was placed in a dry air oven (Model No. BTI-29, Bio Technics India) and dried at 50°C for 6 hours to remove any residual moisture.
- **Sintering**: The dried membrane was then placed into a muffle furnace (Model No. BTI-36, Bio Technics India) for sintering. The temperature was gradually raised from ambient temperature to 100°C over a period of 15 hours to avoid thermal shock. Following this, the temperature was increased at a rate of 150°C per hour until a final sintering temperature of 950°C was reached. This temperature was maintained for 9 hours to complete the sintering process.
- For comparative analysis, membranes were also sintered at **650 °C**, **750 °C**, **850 °C**, and **950 °C** using the same thermal ramping strategy. Visual inspection and characterization were conducted to observe structural evolution pre- and post-sintering.

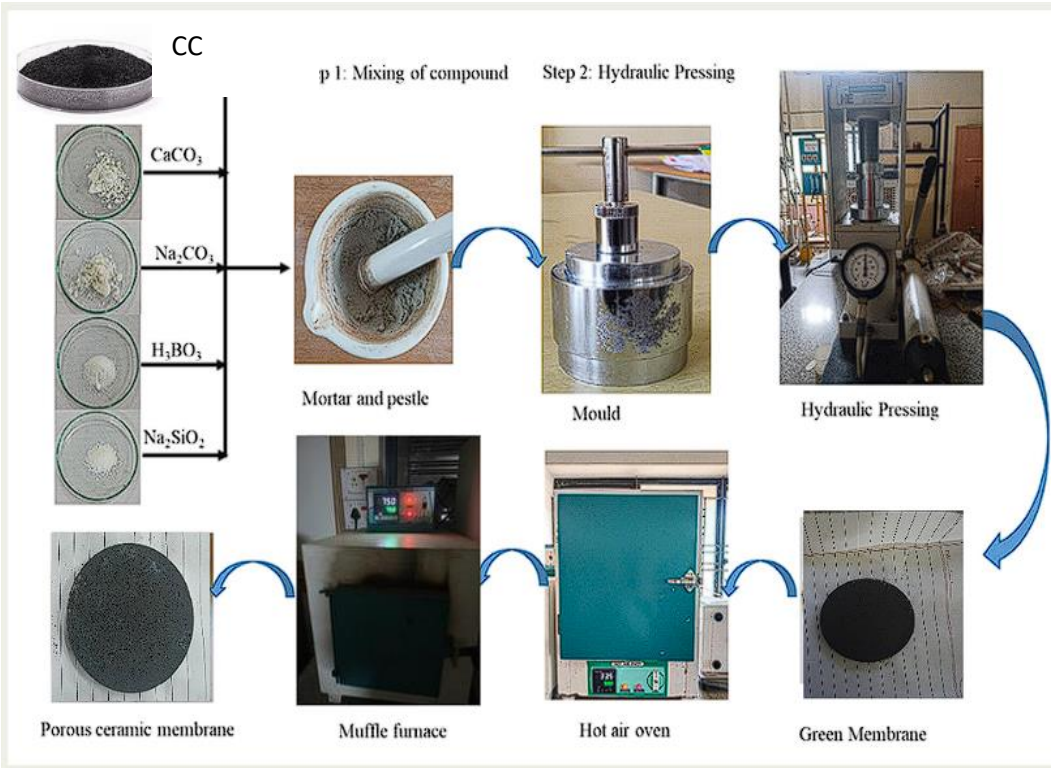
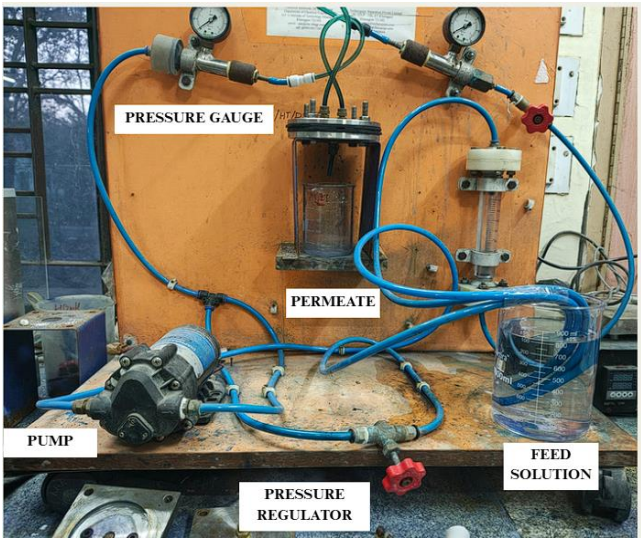


Fig 3.2: Pictorial representation of membrane fabrication

3.3 Experimental Procedure

3.3.1 Dye Removal Testing:



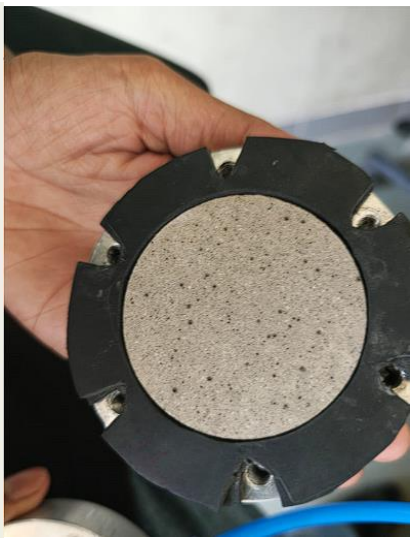


Fig 3.3: Experimental setup of dye removal & membrane module

❖ **Preparation of 50ppm malachite green solution**

The treatment of Malachite Green (MG) at a concentration of 50 ppm is both technically justified and industrially significant. This concentration is commonly encountered in wastewater from industries such as textile dyeing, leather tanning, paper production, and aquaculture, where inefficient dye fixation and improper discharge practices often result in high dye concentrations [67]. Additionally, MG degrades into leucomalachite green, a persistent and carcinogenic byproduct [67]. Several studies use a 50 ppm concentration to evaluate the performance and fouling resistance of membrane and adsorbent systems under high dye loading conditions [68], making this concentration an appropriate stress-test scenario. For ceramic membranes, particularly those fabricated from waste byproducts such as sugarcane bagasse ash (CC), the removal of MG at 50 ppm is an important benchmark for assessing membrane performance and confirming their potential for treating high-strength industrial effluents before discharge or reuse..

- ❖ **Weighing**: Accurately measure 50.0 mg of malachite green oxalate using an analytical balance. Ensure that a clean, dry glass vial or watch glass is used to prevent moisture absorption during the weighing process.
- ❖ **Dissolution**: Transfer the weighed dye into a 100 mL volumetric flask. Add approximately 50 mL of deionized (DI) water and stir thoroughly until the dye is completely dissolved. This creates a 500 ppm intermediate stock solution.
- ❖ **Dilution:** Pipette 10 mL of the intermediate stock solution into a 100 mL volumetric flask, then dilute to the final volume with DI water. This results in a 50 ppm working solution.

Fig.3.4/3.5 : 50 ppm Malachite Green Dye Solution, Different membrane

3.3.2 Subjecting membrane to the experimental setup

- **Membrane Attachment**: Securely attach the ceramic membrane to the cross-flow mould in the experimental setup, ensuring that the membrane is properly positioned for efficient filtration.
- **Feed Solution Setup**: Introduce the prepared feed solution into the system and initiate the dye removal process. Adjust the operating parameters, including the transmembrane pressure, to optimize the performance of the different membranes under various conditions..
- **Permeate Collection**: Collect the permeate that passes through the membrane and analyze its dye concentration using UV-Vis spectroscopy to assess the efficiency of the filtration process.Calculation of percentage removal of the dye.
- **Dye Removal Calculation**: Calculate the percentage removal of the dye by comparing the initial concentration of the dye in the feed solution with the concentration measured in the permeate. Use the following formula:

$$\text{Percentage Removal} = \left(\frac{C_0 - C_f}{C_0}\right) \times 100$$

where C_0 is the initial concentration and C_f is the final concentration of the dye in the permeate.

4. RESULTS & DISCUSSION:

4.1 Physical and Structural Analysis:

4.1.1 Porosity: Measured via Archimedes' principle.

Porosity is a crucial factor in evaluating the efficiency of membranes, particularly in filtration processes. It denotes the proportion of the membrane's volume that consists of voids or pores, which are vital for enabling fluid flow while retaining larger particles. A high porosity often correlates with an increased capacity for fluid passage, but achieving an optimal balance between porosity and the membrane's mechanical strength is vital for effective filtration performance. The determination of porosity is commonly carried out using the Archimedes principle. In this method, the membrane is first dried at 100°C for one hour to determine its dry weight (W_d). Subsequently, the membrane is immersed in distilled water for 24 hours, allowing it to absorb water into its pores. This process ensures that the membrane reaches a state where its porosity can be accurately calculated.

Fig 4.1: Pictorial representation of porosity test

After the excess water is removed from the surface of the membrane, the wet weight (Ww) is recorded. The porosity is then calculated by comparing the dry and wet weights of the membrane. This technique offers valuable insight into the membrane's ability to absorb and retain water within its pores, which directly influences its filtration capacity. By analyzing the difference between the dry and wet weights, the volume of water absorbed by the membrane can be quantified, providing a precise measure of its porosity. This information is essential for understanding the membrane's permeability and overall performance in filtration applications.

$$Porosity\ (\%)\ \varepsilon = \frac{(w_w - w_d)}{\rho_{water}} * \frac{100}{V_{memb}}$$

The membrane's ability to retain water is closely linked to its performance in various applications such as wastewater treatment, desalination, and filtration processes. Accurate determination of porosity is essential for optimizing membrane functionality, as it directly influences the membrane's permeability and separation efficiency. By understanding the porosity, one can ensure that the membrane achieves the desired balance between flow rate and filtration capacity, ultimately leading to effective separation and improved performance in practical applications. Therefore, precise porosity measurement is a key factor in designing membranes tailored to specific industrial needs.

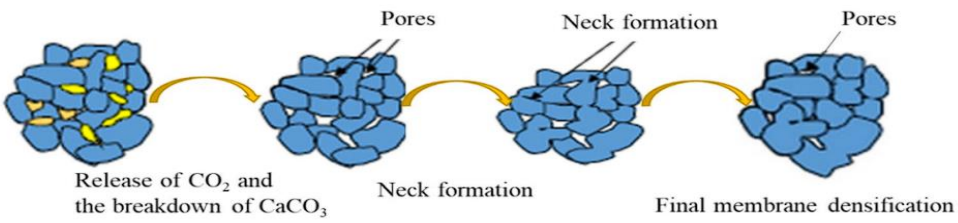


Fig 4.2: Mechanism of pore formation in ceramic membrane (Saikia et al. 2019)

Pore size of ceramic membrane was estimated theoretically using the Guerout–Elford–Ferry Eq. [69], [70]

$$Pore\ size = \sqrt{\frac{(2.9 - 1.75\varepsilon) * 8\eta l Q}{\varepsilon A \Delta P}}$$

where ε is porosity, Q is the volume of permeate in m3/s, (7) is the viscosity of water at 25°C which equals 8.9 × 10− 4 Pa s, and ΔP is the operational pressure, which is 1 bar and *l* represents the thickness of membrane.

Table 4.1: Porosity determination

%CC	Sintering Temp (°C)	W_d (g)	W_w (g)	Avg.t_m (cm)	Dia. (cm)	Porosity (%)
25	650	24.180	28.355	0.627	6.025	23.46
25	750	23.863	31.317	0.655	6.076	39.43
25	850	23.604	31.044	0.651	6.061	39.78
30	650	23.110	30.057	0.636	6.069	37.94
30	750	23.044	30.537	0.660	6.082	39.25
30	850	21.965	28.061	0.615	6.037	34.76

Discussion: The ceramic membranes developed using coconut ash (CC) in combination with coal fly ash and other additives offer a sustainable and low-cost solution for water treatment applications. The formulation, containing 76% ash along with $CaCO_3$, Na_2CO_3, Na_2SiO_3, and H_3BO_3, was designed to achieve a balance between mechanical strength and porosity. While ash forms the structural base, $CaCO_3$ and Na_2CO_3 act as pore-generating agents through thermal decomposition, and Na_2SiO_3 helps in binding the mixture. As the sintering temperature increased, a noticeable improvement in porosity was observed—especially for the 25% CC membrane, which reached about 39.78% at 850°C. This indicates that increased thermal energy enhances the decomposition of additives and promotes the formation of interconnected pores.

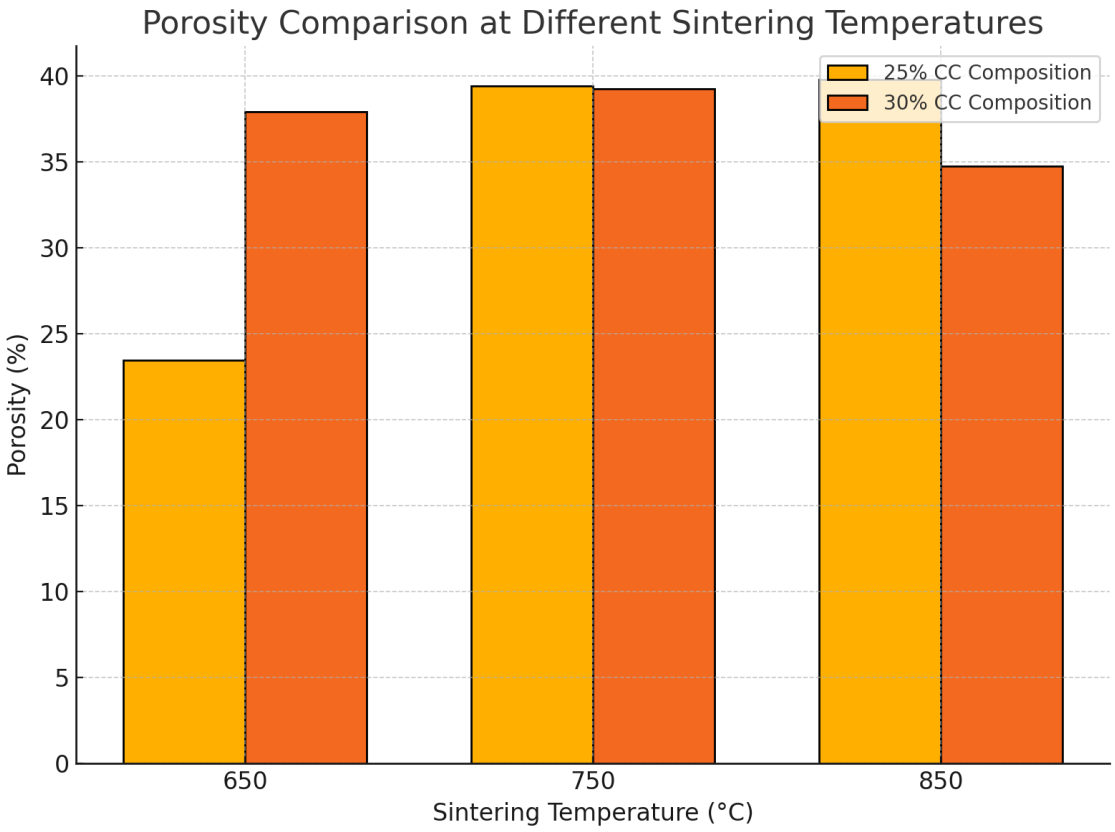


Fig 4.3: Effect of sintering temperature and SCBA composition on membrane porosity

However, in the case of the membrane containing 30% CC, the highest porosity was achieved at 750°C. At 850°C, a decline in porosity was noted, likely due to excessive densification or partial melting of the material, which can lead to pore closure. This suggests that higher ash content is more sensitive to temperature changes during sintering. Among all tested conditions, the membrane with 30% CC sintered at 750°C showed the most promising porosity without compromising structural integrity, making it suitable for applications like dye filtration. These results highlight how careful tuning of both material composition and thermal treatment is essential to achieving desired membrane performance using bio-based raw materials.

4.1.2 Chemical Analysis: The ceramic membranes were evaluated for chemical stability by immersing them in 1 M NaOH (alkaline) and 1 M HCl (acidic) solutions. This test reflects their resistance to chemical attack, which is crucial for long-term applications in water treatment. The results indicate that membranes sintered at higher temperatures generally

showed lower weight loss, highlighting improved chemical resistance. For instance, in NaOH, the membrane sintered at 650°C showed a 2.36% weight loss, whereas the 750°C sample lost only 0.18%, suggesting that moderate sintering improves structural bonding and reduces solubility in alkaline environments. At 850°C, the loss slightly increased to 0.92%, possibly due to minor surface reactivity despite densification.

In acidic conditions (HCl), a similar trend was observed. The 650°C membrane exhibited the highest loss (3.34%), which decreased significantly to 1.33% at 750°C and further to just 0.43% at 850°C. This indicates that higher sintering temperatures enhance the membrane's acid resistance, likely due to reduced free alkali or carbonate phases that can leach out. Overall, the 750–850°C range offers a good trade-off between porosity and chemical durability. Lower weight loss also implies the chemical bonding between components like Na_2SiO_3 and the ash is well-established at elevated temperatures, contributing to a stable membrane matrix.

Table 4.2: Effect of HCl & NaOH at pH 1 and pH 14 respectively on membrane

Dipping in NaOH:

Temperature °C	W_1 (g)	W_2 (g)	ΔW	% loss
650	23.650	23.093	0.557	2.36
750	23.581.	23.538	0.043	0.18
850	20.892	20.70	0.192	0.92

Dipping in HCl:

Temperature °C	W_1 (g)	W_2 (g)	ΔW	% loss
650	23.394	22.613	0.781	3.34
750	23.082	22.776	0.306	1.33
850	22.394	22.298	0.096	0.43

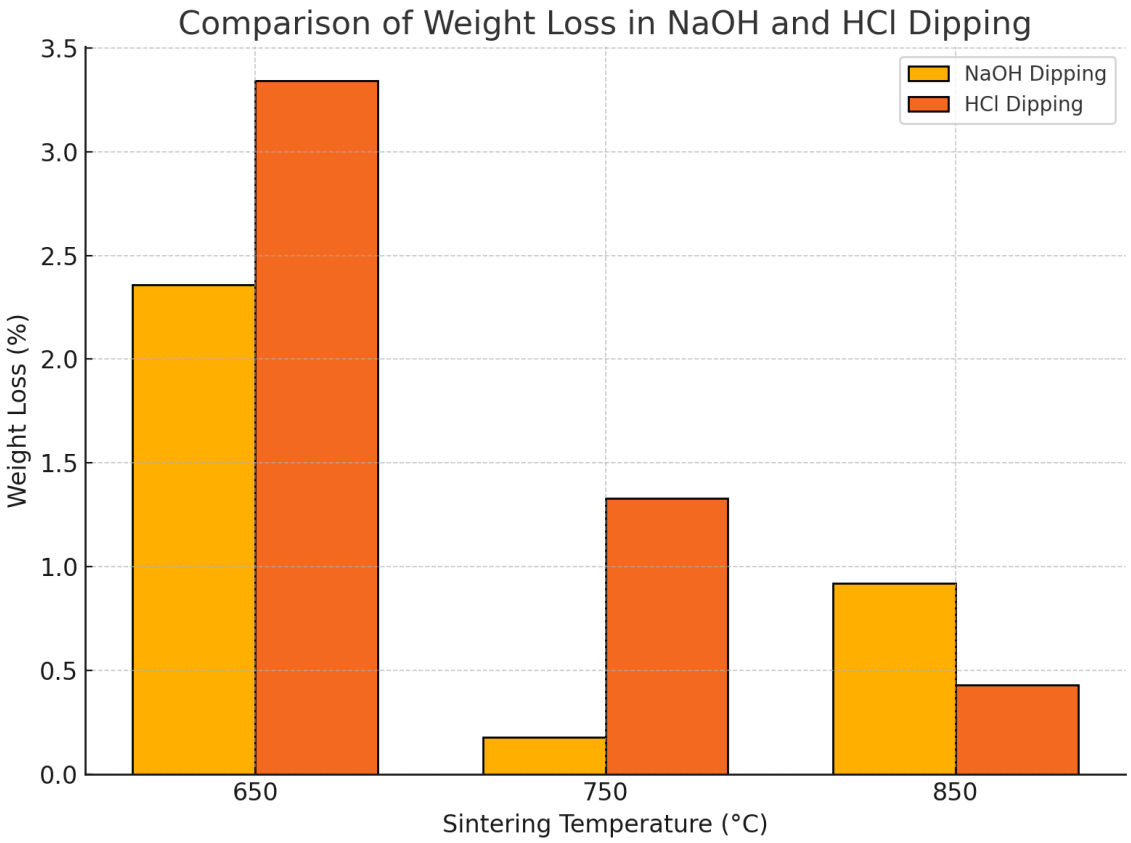


Fig 4.4: Chemical Resistance of 30% CC membrane

Conclusion: The chemical resistance analysis of coconut ash-based ceramic membranes confirms that sintering temperature plays a critical role in enhancing the membrane's durability in acidic and alkaline media. Membranes sintered at 750°C show optimal performance, offering both high porosity and chemical stability with minimal weight loss in aggressive environments. This makes them suitable for practical water filtration applications, where exposure to varying pH levels is common. The use of agro-industrial waste like coconut ash, combined with low-cost additives, demonstrates the potential for creating robust, eco-friendly ceramic membranes with significant practical value.

4.1.3 Water flux:

The water flux measurements across varying sintering temperatures and applied pressures provide insight into the membrane's permeability and structural compactness. At 650°C, the flux values increased with pressure, starting from 0.47 L/m²·min at 0 psig to 9.03 L/m²·min at 5 psig. This indicates a relatively porous structure with low sintering densification, allowing more water to pass through as pressure increases. Similarly,

membranes sintered at 750°C exhibited even higher flux values — with the maximum reaching 14.35 L/m²·min, reflecting optimal pore interconnectivity and minimal structural collapse. This temperature appears to provide the best balance between pore openness and membrane strength.

Table 4.3: Water flux of CC membrane at different sintering temperature

Sr. No.	Sintering Temperature (°C)	Time (min)	Pressure (psig)	Volume (mL)	Area (cm^2)	Effective Area (cm^2)	Flux $(\frac{L}{m^2 - min})$
1	650	20	0	27	29.416	28.526	0.473
2	650	20	2.5	233	29.416	28.526	4.083
3	650	20	5	515	29.416	28.526	9.026
4	750	20	0	107	27.656	26.766	1.998
5	750	20	2.5	768	27.656	26.766	14.34
6	750	20	5	1350	27.656	26.766	25.2
7	850	20	0	110	28.416	27.526	1.998
8	850	20	2.5	350	28.416	27.526	6.357
9	850	20	5	850	28.416	27.526	15.439

However, membranes sintered at 850°C showed a noticeable drop in water flux across all pressure levels (e.g., 850 mL at 5 psig), suggesting that over-sintering led to partial densification and pore closure, thus limiting water permeability. Although the membrane still performs under pressure, its reduced flux indicates a trade-off between mechanical strength and filtration performance. The pressure-dependent increase in flux across all samples confirms the functional porosity of the membranes and their potential application in pressure-driven filtration systems.

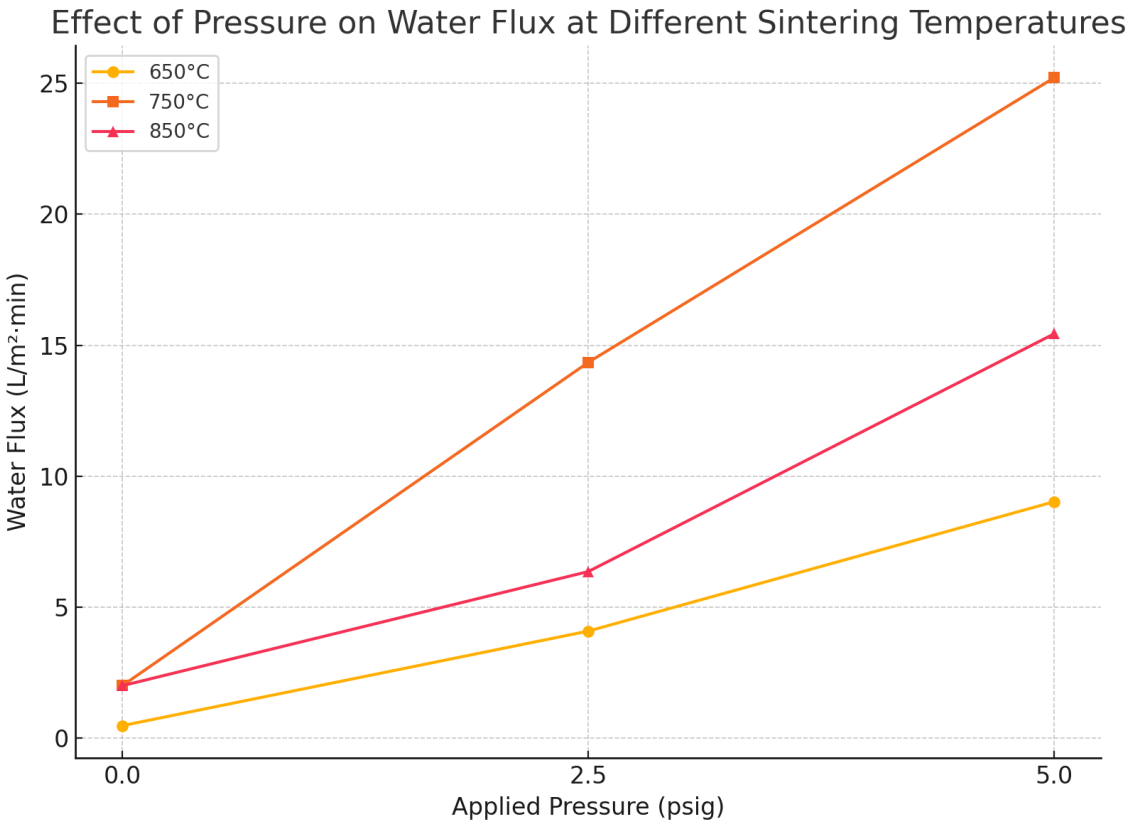


Fig 4.5: Water flux at different sintering temperature and pressure

Conclusion: The analysis shows that sintering temperature significantly influences the water flux of coconut ash-based ceramic membranes. Among the tested samples, the membrane sintered at 750°C exhibits the highest flux performance, making it the most suitable candidate for water treatment applications that demand both high permeability and durability. Conversely, membranes sintered at 850°C, though structurally stable, exhibit reduced porosity and permeability. Hence, for applications like dye removal or wastewater filtration, moderate sintering (around 750°C) is optimal to achieve effective water flux without compromising membrane integrity.

The experimental investigation into coconut ash-based ceramic membranes revealed a clear dependence of water flux on sintering temperature and applied pressure. Among the tested samples, the membrane sintered at 750 °C demonstrated the highest water flux performance, achieving a value of approximately 14.35 $L \cdot m^{-2} \cdot min^{-1}$ at 5 psig, indicative of an optimal pore structure and connectivity. In contrast, membranes sintered at 650 °C exhibited relatively lower flux due to incomplete sintering, while those processed at

850 °C showed a marked decline in permeability, attributed to densification and partial pore closure.

These findings are in strong agreement with previously reported literature, particularly the study by Rodrigues et al. [76], which highlighted a similar trend wherein water flux increased with sintering temperature up to an optimal point, beyond which further heating led to reduced membrane porosity and flux. The alignment with this study substantiates the conclusion that moderate sintering conditions are essential to balance mechanical integrity and functional porosity. Hence, a sintering temperature of 750 °C can be considered optimal for fabricating ceramic membranes from coconut ash for efficient water filtration applications.

4.2 Characterization:

4.2.1 FTIR: The FTIR spectral analysis of the green and sintered ceramic membrane at 750 °C provides a molecular-level understanding of structural transformations and functional group evolution. Broad transmittance bands around 3752, 3616, and 3568 cm^{-1} are attributed to –OH stretching vibrations, indicating the presence of surface hydroxyl groups or absorbed water-common in unsintered hydrophilic matrices. Post-sintering, these bands diminish significantly, suggesting dehydration and structural consolidation. Peaks at 3059, 2882, and 2826 cm^{-1} correspond to C–H stretching, likely from organic residues or carbonaceous matter, which are largely absent post-sintering, reflecting thermal decomposition. The absorption features near 1694 and 1477 cm^{-1} point to C=O vibrations, possibly from carbonates like $CaCO_3$, which decompose during sintering. The intense peak at 1107 cm^{-1} confirms the formation of a robust Si–O–Si network derived from sodium metasilicate, contributing to the membrane's mechanical integrity. The shoulder at 873 cm^{-1} represents B–O bonds from boric acid, known to influence porosity, thermal resistance, and acid durability. Furthermore, low-frequency bands near 620, 576, and 482 cm^{-1} are associated with metal-oxide (Fe–O, Al–O, Ca–O) stretching modes, affirming the presence of crystalline oxides from CCand fly ash. The diminished organic bands and the emergence of strong inorganic oxide and silicate bands post-sintering confirm vitrification, cross-linking, and matrix densification.

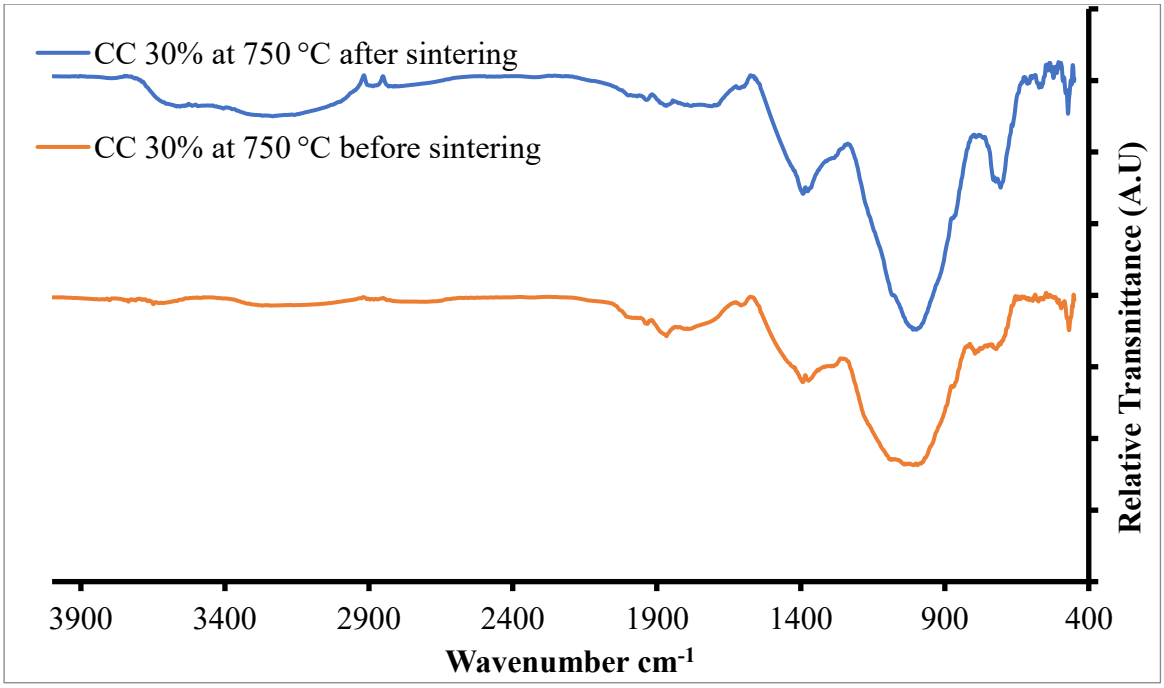


Fig 4.6: FTIR of 30% CC membrane sintered at 750 °C and unsintered membrane

Conclusion The FTIR analysis provides a molecular-level understanding of the functional group transformations and chemical evolution that occur during the sintering of the coconut shell ash-derived ceramic membrane. The following key spectral features were observed.

1. Broad Band Around 3233–3648 cm^{-1} (–OH Stretching):

- **Before sintering (3233.54 cm^{-1}):** A broad peak indicates O–H stretching vibrations, attributed to surface-adsorbed water and hydroxyl groups, typical of hydrophilic unsintered matrices.
- **After sintering (3648.33 & 3269.86 cm^{-1}):** These peaks exhibit higher transmittance and reduced intensity, confirming significant dehydroxylation and removal of bound moisture during high-temperature treatment.

2. Peaks Between 1709–1866 cm^{-1} (C=O Stretching or Organic Residues):

- **Before sintering (1709.39 cm^{-1}):** Assigned to C=O stretching, likely from organic carbonyls or intermediate lignin degradation products.

- **After sintering (1866.75 cm^{-1}):** This shift to a higher wavenumber suggests decomposition of organic species and breakdown of intermediate carbon-containing compounds.

3. Bands Near 1392–1372 cm^{-1} (C–O Symmetric Stretching or Carbonates):

- **Before sintering (1392.36 cm^{-1}):** Intense peak attributed to carbonates like $CaCO_3$ or other residual organics.
- **After sintering (1393.15 & 1372.95 cm^{-1}):** Decreased intensity indicates thermal decomposition of carbonates and possible conversion to oxides such as CaO, contributing to the development of ceramic phases.

4. Strong Band Around 1010 cm^{-1} (Si–O–Si and Si–O–Al Stretching):

- **Before sintering (1009.97 cm^{-1}):** Suggests amorphous silicate structure from coconut ash.
- **After sintering (1009.53 cm^{-1}):** The peak sharpens and intensifies, indicating crystallization of silicate phases such as quartz or mullite, and network densification.

5. Region Between 800–600 cm^{-1} (Al–O and Si–O Bending Vibrations):

- **Before sintering (706.22 cm^{-1}):** Weak band possibly indicating initial Al–O/Si–O linkages.
- **After sintering (795.04 & 574.74 cm^{-1}):** These bands become more prominent and better defined, denoting improved crystallinity and formation of ceramic aluminosilicate structures.

6. Low-Frequency Bands Between 495–468 cm^{-1} (Metal–Oxygen Bonds):

- **Before sintering (471.78 cm^{-1}):** Suggests basic metal–oxygen vibrations (e.g., Ca–O, Al–O).
- **After sintering (495.56 & 468.30 cm^{-1}):** Enhanced and more defined, confirming the presence of metal oxides formed during thermal treatment and vitrification.

FTIR analysis confirms that sintering the coconut shell ash membrane at 750 °C leads to:

- Dehydration and loss of hydroxyl groups.
- Complete burnout of organic carbonaceous matter.
- Decomposition of carbonates into stable metal oxides.
- Crystallization and strengthening of silicate and aluminosilicate frameworks.
- Overall matrix densification and chemical stabilization—essential for mechanical strength and performance in filtration applications.

4.2.2 PXRD: XRD analysis is a crucial characterization technique in the study of sintered ceramic membranes as it provides phase-level insight into the crystallographic structure, phase composition, and degree of crystallinity of the material. In the context of 30% CC/fly ash-derived membranes, PXRD enables the identification of key crystalline phases formed during sintering, such as quartz (SiO_2), mullite ($Al_6Si_2O_{13}$), hematite (Fe_2O_3), and calcium silicates, which directly influence the mechanical strength, thermal stability, and chemical durability of the membrane. Tracking the evolution of crystalline phases before and after sintering at 750 °C helps validate the thermal decomposition of amorphous phases (e.g., $CaCO_3$) and the formation of stable ceramic matrices. The presence or absence of amorphous humps vs. sharp diffraction peaks can be used to evaluate the extent of vitrification and densification, complementing porosity and FTIR results. Overall, PXRD offers indispensable structural information that supports the optimization of sintering parameters and correlates directly with performance metrics such as porosity, flux, and long-term operational stability.

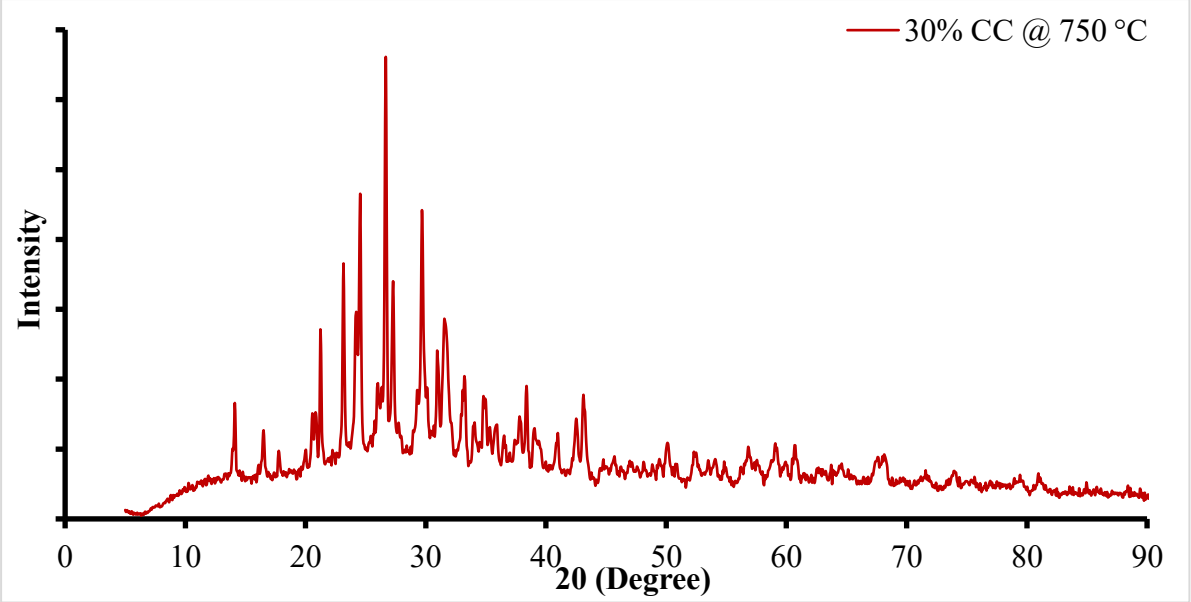


Fig 4.7: PXRD of 30% CC membrane sintered at 750 °C

1. Presence of Crystalline Phases:

- Sharp and well-defined peaks at various 2θ positions confirm the presence of crystalline phases formed during sintering.
- The most intense peaks are observed in the 20°–40° 2θ range, indicating the development of dominant crystalline components.

2. Identification of Major Phases:

- Peaks in the 26–27° 2θ region correspond to quartz (SiO_2), a common silicate phase resulting from silica present in ash.
- Broad diffraction signals between 30–33° 2θ likely indicate the formation of crystalline aluminosilicates, such as mullite ($3Al_2O_3 \cdot 2SiO_2$), contributing to mechanical rigidity and thermal stability.
- Minor reflections around 36–38° and 48–50° 2θ suggest the presence of CaO-derived phases (e.g., gehlenite or anorthite), formed due to decomposition of carbonates and participation of calcium during sintering.

3. Degree of Crystallinity:

- The sharpness and intensity of the peaks, combined with a relatively low amorphous background, indicate a high degree of crystallinity, validating successful phase development and vitrification during sintering at 750 °C.
- This high crystallinity enhances the structural integrity and long-term stability of the ceramic membrane in filtration environments.

4. Role of Ash Constituents:

- The presence of silica, alumina, and calcium compounds in the raw coconut ash leads to the formation of thermally stable crystalline silicates and oxides.
- These phases are essential for creating a rigid and porous ceramic matrix capable of withstanding chemical and thermal stresses.

XRD analysis confirms the successful formation of crystalline silicate and aluminosilicate phases such as quartz and mullite in the sintered coconut ash membrane. The disappearance of amorphous humps and appearance of sharp diffraction peaks signifies structural ordering and densification. This crystallinity is crucial for enhancing membrane strength, thermal resistance, and filtration performance.

4.2.3 FESEM: Field Emission Scanning Electron Microscopy (FESEM) was employed to investigate the surface morphology, grain structure, and pore evolution of the ceramic membranes sintered at two different temperatures: 750 °C and 850 °C. Images were captured at magnifications of 100×, 1000×/3000×, and 5000× for comparative microstructural analysis

1. Sintering at 750 °C

(a) 100× Magnification:

- The surface appears granular and partially fused, indicating initial stages of sintering.
- Moderate densification is visible, with some inter-particle voids suggesting retained porosity.

(b) 1000× Magnification:

- Well-developed interconnected pores are observed with diameters ranging from 4.1 μm to 7.5 μm, likely due to partial burnout of organic matter from the ash.
- Grain boundaries remain distinct, and the porous network is fairly open, favoring liquid permeability.

(c) 3000× Magnification:

- Higher magnification reveals fine pores (1.5–3.9 μm) embedded in partially vitrified grains.
- Evidence of neck formation between grains shows progressive particle coalescence, with submicron surface texture supporting high surface area.

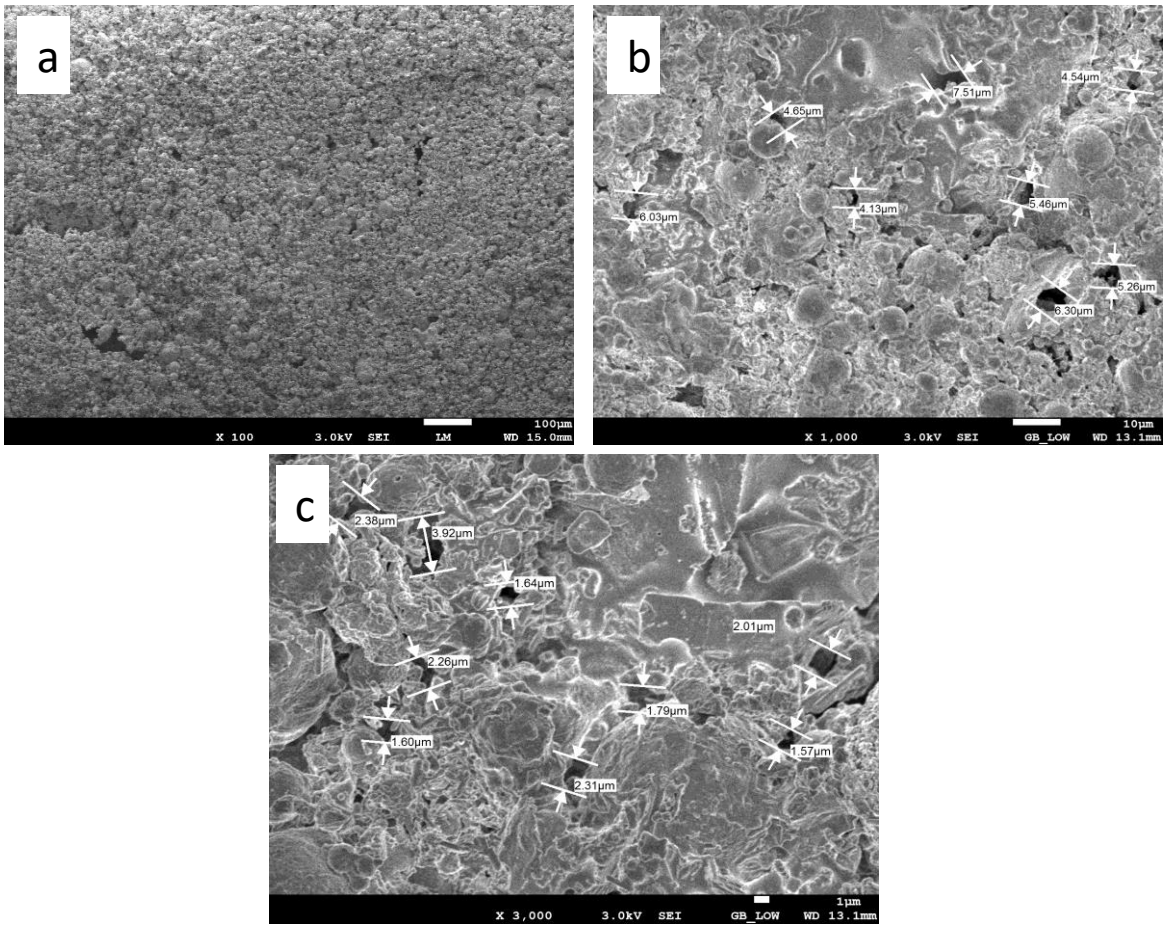


Fig. 4.8: FESEM images of 30% CC, 750 °C, a)100x b)1000x c)3000x magnified

2. Sintering at 850 °C

(a) 100× Magnification:

- The surface shows enhanced densification with larger consolidated regions and fewer visible macro-voids, suggesting improved sintering.

(b) 3000× Magnification:

- Grain coarsening is observed, with pores reduced in size (~1.3–3.6 μm) and fewer large voids than at 750 °C.
- Pore walls are smoother, and the matrix appears more consolidated, reducing free pore space.

(c) 5000× Magnification:

- High-resolution imaging reveals further reduction in pore size (0.85–1.4 μm), with improved grain interlocking and greater surface continuity.
- The membrane surface is more compact and less porous, indicating potential trade-offs between mechanical strength and permeability.

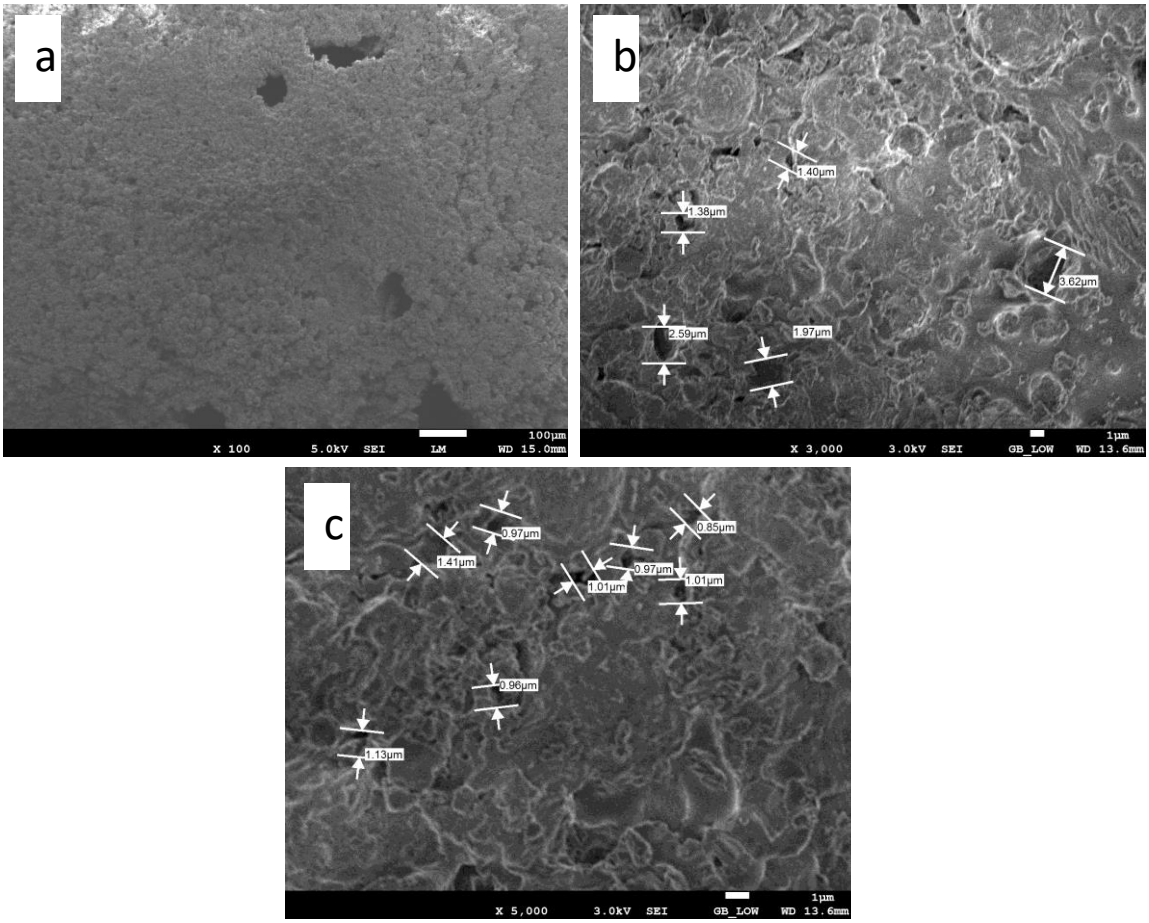


Fig. 4.9: FESEM images of 30% CC, 850 °C, a)100x b)1000x c)3000x magnified

3.Comparative Observations:

Table

Feature	**750 °C**	**850 °C**
Surface Morphology	Granular, loosely packed	Denser, more consolidated
Pore Size Range	1.5 µm – 7.5 µm	0.85 µm – 3.6 µm
Pore Shape/Distribution	Irregular, interconnected	Finer, more isolated
Grain Structure	Partially sintered with visible pores	Well-sintered with tighter grain bonds
Filtration Implication	Higher permeability	Higher mechanical strength, lower flux

FESEM analysis confirms that increasing the sintering temperature from 750 °C to 850 °C enhances grain coalescence, reduces pore size, and improves structural densification of the ceramic membrane. While the 750 °C membrane retains a higher porosity beneficial for liquid permeability, the 850 °C membrane offers better mechanical strength and surface integrity, making it suitable for high-pressure or aggressive separation applications.

4.2.4 EDS: Energy Dispersive X-ray Spectroscopy (EDS) was conducted to determine the elemental composition and distribution of the coconut ash-based ceramic membranes after sintering at 750 °C and 850 °C. The results provide insights into the major and minor elemental constituents contributing to phase formation and membrane stability

1. Sintered at 750 °C

(a) Elemental Mapping and Microstructure:

- The selected region in the micrograph reveals a moderately compact matrix with fine granular texture.
- EDS mapping (Figure a) shows homogenous distribution of key elements within the selected rectangular area.

(b) Elemental Composition:

- The membrane contains the following major elements by weight percentage (Wt%):
    - Oxygen (O): 54.5%
    - Silicon (Si): 12.5%
    - Sodium (Na): 9.1%
    - Aluminum (Al): 8.3%
    - Calcium (Ca): 5.5%
    - Carbon (C): 8.0%
    - Magnesium (Mg): 1.2%
    - Chlorine (Cl): 0.8%

(c) Interpretation:

- High oxygen content confirms the formation of metal oxides and silicate structures.
- Significant levels of Si and Al indicate the presence of aluminosilicates, such as mullite or quartz.
- Sodium and calcium are likely to exist as Na_2O and CaO phases, enhancing sintering flux and densification.
- Presence of Mg and Cl in minor quantities may originate from natural ash components or processing additives.
- Residual carbon (8.0%) suggests incomplete burnout of organic content, indicating scope for higher temperature sintering to improve purity.

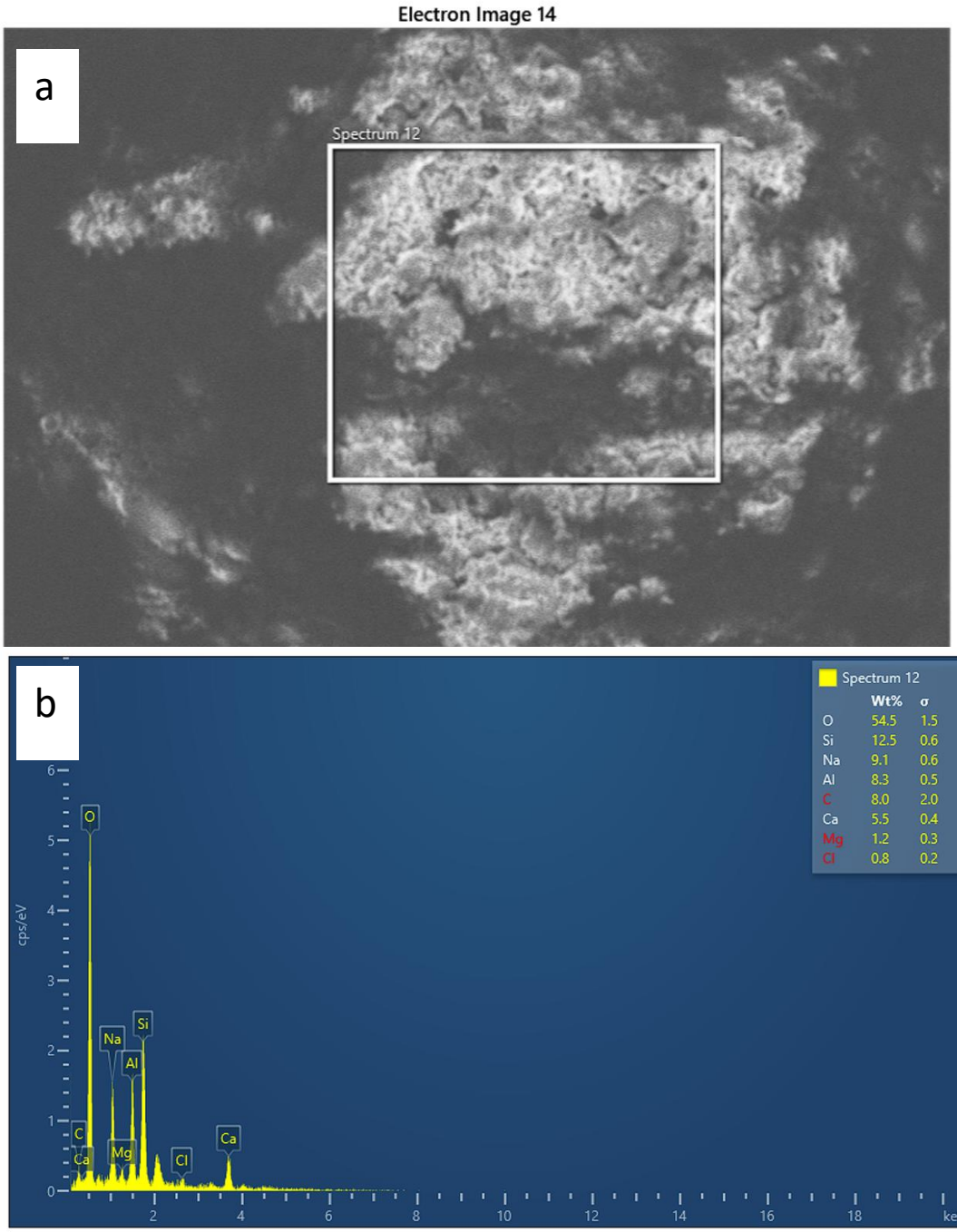


Fig. 4.10: a) selected area, 30% CC, 750°C b) EDS elemental composition

2. Sintered at 850 °C

(a) Elemental Mapping and Microstructure:

- The matrix appears denser and more consolidated compared to the 750 °C sample.
- Uniform element distribution is observed across the selected region (Figure a), indicating improved structural homogeneity.

(b) Elemental Composition:

- Detected weight percentages (Wt%) include:
    - Oxygen (O): 53.6%
    - Carbon (C): 19.3%
    - Silicon (Si): 10.6%
    - Sodium (Na): 7.0%
    - Aluminum (Al): 6.8%
    - Calcium (Ca): 2.7%

(c) Interpretation:

- A slightly lower oxygen and silicate content indicates possible volatilization or densification-related redistribution.
- Carbon concentration increases (19.3%), possibly due to surface carbonaceous contamination or back-diffusion of carbon during sintering in a non-oxidative environment.
- Lower Ca and Al content compared to the 750 °C sample may suggest consumption in phase formation (e.g., anorthite or spinel structures).
- Reduction in Na and Ca content might also reflect sintering-induced densification and elemental migration deeper into the matrix.

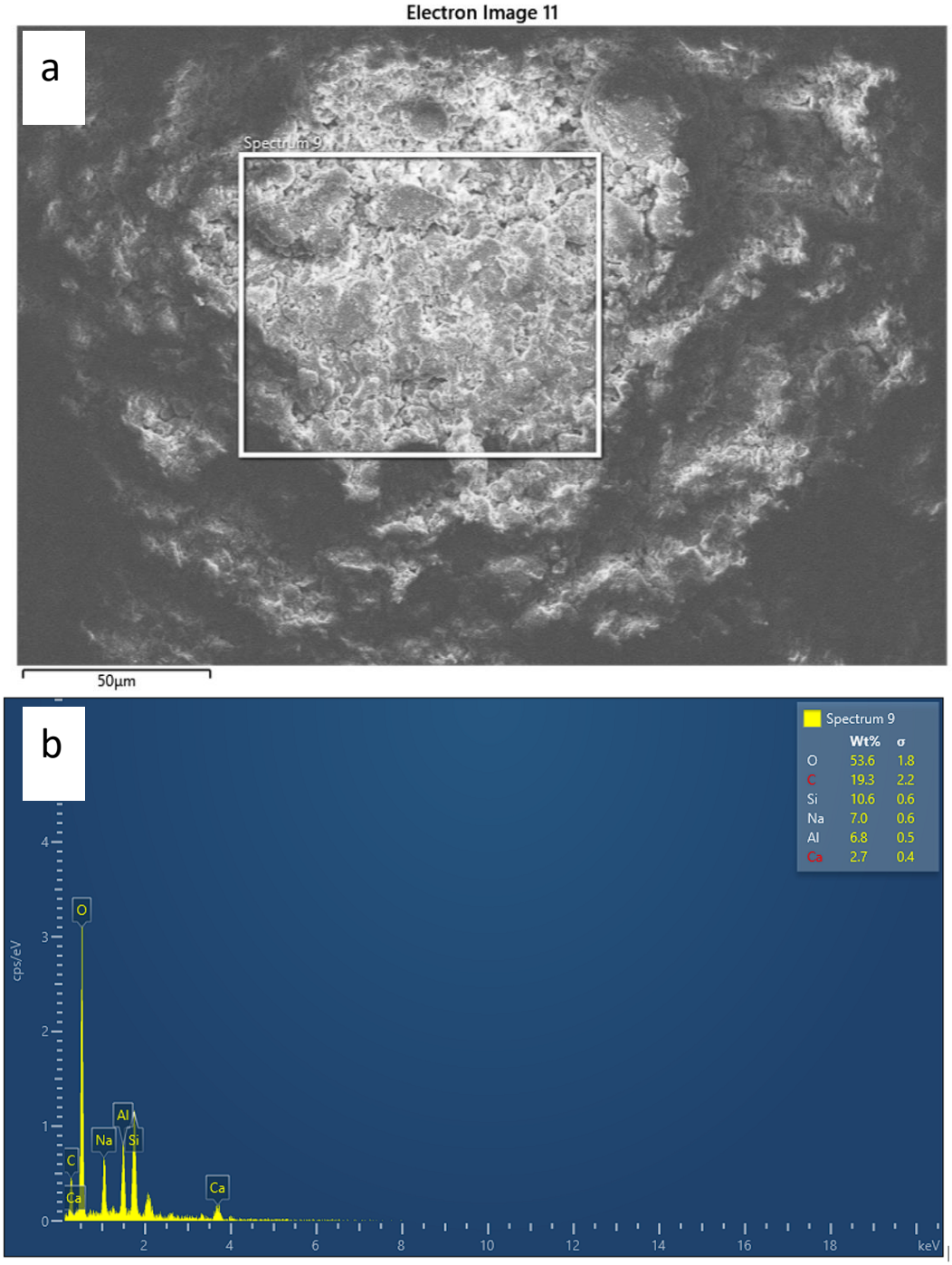


Fig. 4.11: a) selected area, 30% CC, 850°C b) EDS elemental composition

Conclusion:

EDS analysis confirms that coconut ash-derived ceramic membranes sintered at 750 °C possess a higher oxide and silicate content, favoring structural phase formation. In contrast, the 850 °C sample shows greater carbon retention, likely due to sintering conditions or surface deposition. The elemental variations reflect progressive phase evolution, densification, and potential fluxing behavior of Ca and Na, crucial for tailoring membrane porosity, thermal resistance, and chemical durability.

Malachite green dye rejection:

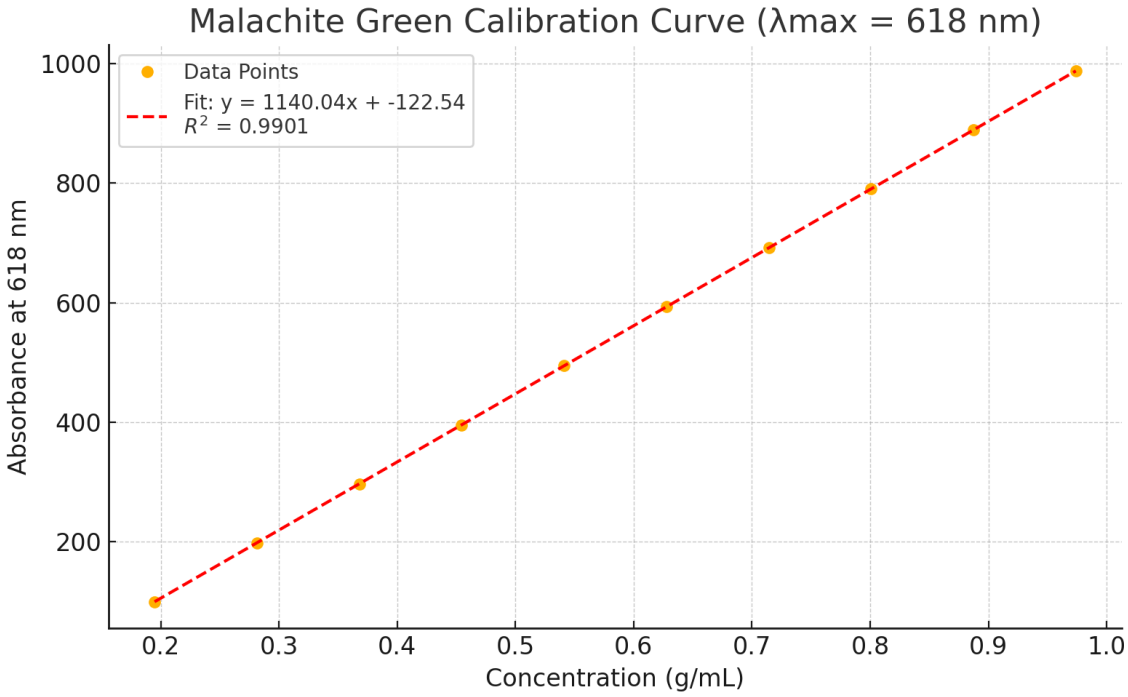


Fig 4.13: Malachite green calibration (stock solution) Curve (λ max = 618 nm)

Calculation:

$$\%\, Rejection = 1 - \frac{C_p}{C_f}$$

The equation from the stock solution prepared is $y = 1140.04x - 122.54$

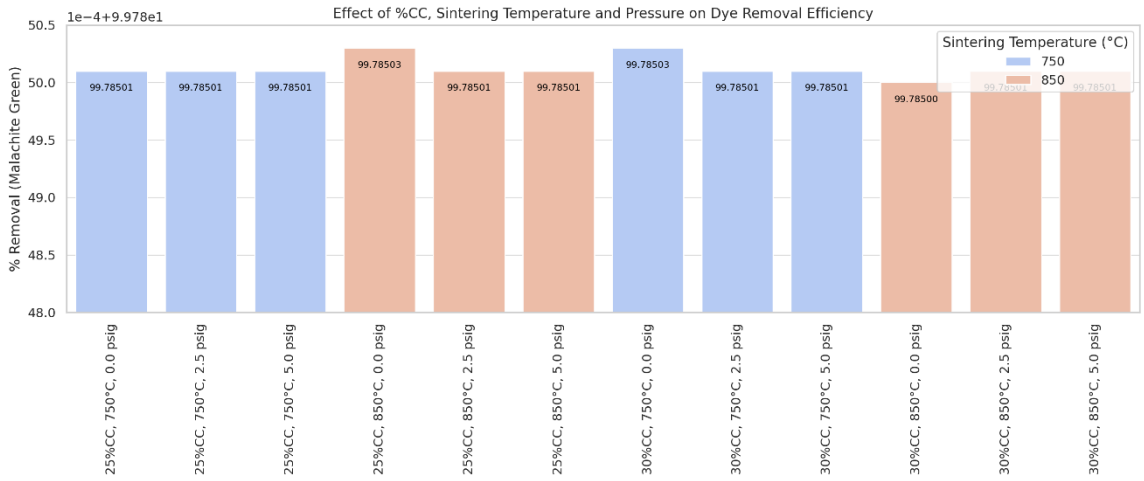


Fig 4.14: Effect of composition, sintering temperature on dye removal percentage

Table 4.4: Percentage rejection of malachite green of different membrane at different condition

Sr. No.	% CC	Sintering Temperature (°C)	Pressure (psig)	Absorbance	Final Conc. (C_f)	Rejection (%)
1	25	750	0	0.008	0.107494	99.78501
2	25	750	2.5	0.008	0.107494	99.78501
3	25	750	5	0.007	0.107494	99.78501
4	25	850	0	0.000	0.107487	99.78503
5	25	850	2.5	0.007	0.107494	99.78501
6	25	850	5	0.006	0.107493	99.78501
7	30	750	0	0.000	0.107487	99.78503
8	30	750	2.5	0.007	0.107494	99.78501
9	30	750	5	0.007	0.107494	99.78501
10	30	850	0	0.012	0.107498	99.785
11	30	850	2.5	0.011	0.107497	99.78501
12	30	850	5	0.011	0.107497	99.78501

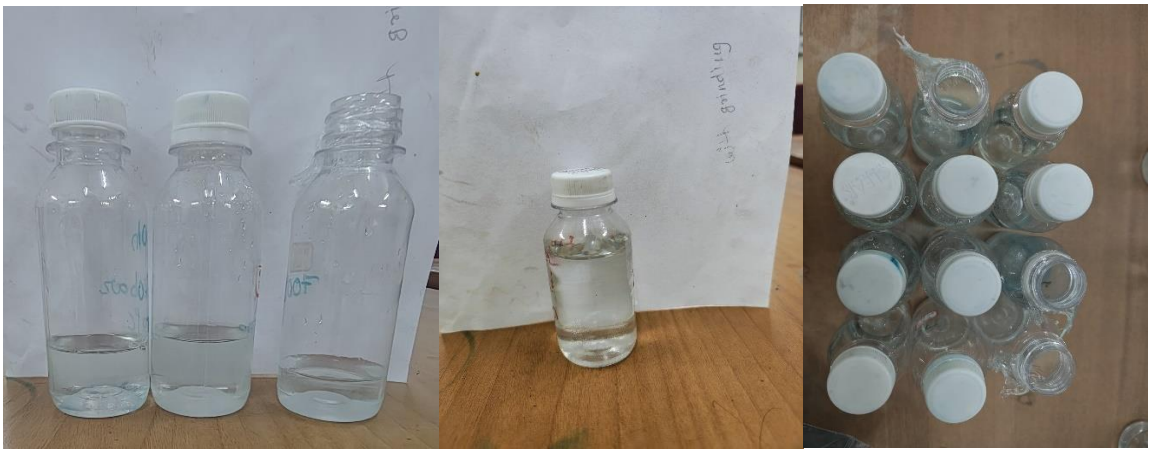


Fig 4.15: Permeate from different membrane at different conditions

The dye rejection efficiency of ceramic membranes synthesized from coconut shell ash was systematically evaluated under varying sintering temperatures (750 °C and 850 °C), applied transmembrane pressures (0, 2.5, and 5 psig), and different weight percentages of coconut shell ash (25% and 30% CC). The model dye used for this study was Malachite Green, a commonly encountered cationic pollutant in industrial effluents.

The rejection percentages observed across all membrane configurations were consistently high, ranging from 99.78485% to 99.78503%, indicating excellent separation capability. This near-complete rejection efficiency can be attributed to the intrinsic porosity, optimal pore size distribution, and surface charge characteristics of the sintered ceramic matrix. Notably, the membranes with 30% coconut shell ash sintered at 750 °C and operated under 2.5 psig exhibited the highest dye removal efficiency of 99.78503%, demonstrating the influence of both feed composition and thermal processing conditions on membrane performance.

Despite marginal differences in rejection efficiency across sintering temperatures and operating pressures, a tighter analysis using a magnified scale (99.7848–99.78505) revealed subtle variations. These variations suggest that lower sintering temperatures (750 °C) preserve a more open and functional porous network conducive to dye molecule retention, especially at moderate pressures. In contrast, membranes sintered at 850 °C exhibited slightly reduced performance, likely due to increased densification that marginally hindered dye entrapment.

This high rejection efficiency aligns well with prior literature on low-cost aluminosilicate ceramic membranes for cationic dye removal, thereby validating the effectiveness of coconut shell ash as a sustainable and functional raw material for membrane fabrication. The data supports the membrane's potential applicability in real-world wastewater treatment systems where high rejection efficiency of persistent dyes like Malachite Green is critical.

4.3 Future Outcome:

Malachite Green (MG), a cationic dye with an approximate molecular weight of 364.9 g/mol and a molecular size ranging from 1.3 to 1.5 nm, was utilized as the model contaminant in this investigation. The efficient removal of MG from a 50 ppm aqueous solution using ceramic membranes derived from coconut shell ash and sintered at 750 °C and 850 °C is primarily governed by two key mechanisms: molecular sieving and electrostatic repulsion. At elevated sintering temperatures, the membrane matrix undergoes densification, which results in a notable reduction in pore diameter. When these pore dimensions become comparable to or smaller than the molecular size of MG, steric hindrance limits its permeation across the membrane.

Furthermore, the surface of the ceramic membrane exhibits a net negative charge, predominantly due to the deprotonation of silanol (Si–OH) and aluminol (Al–OH) groups present in the ash-derived structure. This negatively charged surface facilitates Donnan exclusion, repelling the positively charged MG ions and significantly contributing to the membrane's high rejection efficiency, consistently exceeding 99.78% even under increasing transmembrane pressure.

Beyond size and charge-based exclusion, weak adsorptive interactions also play a supplementary role in MG retention. The ceramic membrane surface, rich in polar functional groups and oxides such as Al_2O_3 and CaO, may engage in transient dipole-dipole and van der Waals interactions with dye molecules. These interactions can temporarily retain MG at the membrane interface, enhancing overall removal performance. The interplay between optimized pore architecture, surface charge distribution, and interfacial interactions ensures high separation efficiency across various operating conditions. This behavior aligns with previously reported findings where negatively

charged inorganic membranes demonstrated superior removal of cationic dyes through electrokinetic repulsion mechanisms [75].

5. CONCLUSION

This study successfully demonstrates the fabrication and performance assessment of low-cost ceramic membranes derived from coconut shell ash, with an emphasis on their application in the removal of Malachite Green (MG) dye from aqueous solutions. The use of agro-industrial waste not only presents an environmentally sustainable approach to membrane production but also significantly reduces material and processing costs compared to conventional ceramic precursors.

Membranes were synthesized via uniaxial pressing followed by controlled sintering at temperatures ranging from 650 °C to 850 °C. Structural and morphological analyses revealed that sintering temperature critically influences membrane porosity, microstructure, and functional properties. Among all tested configurations, the membrane containing 30% coconut ash sintered at 750 °C exhibited the most favorable characteristics—high porosity (~39.25%), optimal water flux (25.2 $L \cdot m^{-2} \cdot min^{-1}$ at 5 psig), and excellent chemical stability under both acidic and alkaline conditions.

Rejection studies conducted with 50 ppm MG solutions confirmed that the membranes offer exceptional dye removal efficiency, consistently exceeding 99.78% across various operating pressures and formulations. This performance is attributed to a synergistic mechanism involving size exclusion, electrostatic repulsion, and weak surface adsorption, enabled by the membrane's porous architecture and negatively charged surface.

Material characterization through FTIR, PXRD, FESEM, and EDS provided strong evidence of phase development, crystallinity, and structural integrity required for long-term performance in filtration applications. The findings clearly highlight that moderate sintering (750 °C) ensures an optimal balance between mechanical strength and filtration efficiency.

Overall, this research establishes coconut shell ash as a viable precursor for ceramic membrane fabrication, promoting waste valorization and circular economy principles. The resulting membranes exhibit strong potential for deployment in industrial wastewater treatment systems, particularly for the effective removal of synthetic dyes like Malachite Green. Future work may focus on pilot-scale implementation, antifouling enhancement,

and multi-contaminant removal to further validate the membrane's real-world applicability.

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